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Computer Simulation of Martensitic Transformation Under Stress

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Abstract: This paper reports a computer simulation of martensitic transformation in both Cu-Zn-Al and Fe-Ni-C alloys by a 2-dimensional model.

The influence of the imposed stress on the mechanical behaviour, the transformation kinetics, and the microstructure are illustrated for Cu-Zn-Al alloy. The study of transformation kinetics shows the following features: (1) in case of stress-free transformation, both alloys transform in burst, the transformation will go directly to near completion once a band can overcome the energy barrier. (2) the transformation temperature range spreads monotonically as the load is increased from 0 to 500 MPa. (3) M_f is nearly the same for the different tested loading conditions. Even if the assumption of elastic accommodation is not realistic for Fe-Ni-C alloy, we can analyse the influence of the volume change associated with the transformation on the local stress level and on the behaviour of the alloy. This volume change has no notable influence at the earlier stage of the transformation. It becomes larger as the transformed fraction increases.

1. INTRODUCTION

The effect of stress on martensitic transformation results in modifications of the transformation kinetics, of the martensite morphologies and of the mechanical behaviour. Whereas the pseudoelastic behaviour of the shape memory alloys has been largely studied, the analysis of the mechanical behaviour of iron-based alloys during their phase transformation has received less interest. Particularly, the different mechanisms at the origin of transformation plasticity and the role of the internal stresses has been little discussed [1,2]. Computer simulation may help us to understand better the effect of the local stresses with regard to the externally applied stress on the microstructural evolution and on the mechanical behaviour. Several investigators have contributed a lot in this area using different methods such as molecular dynamics [3], self consistent schemes [4] and finite elements (FEM) [5-9]. Whereas molecular dynamics simulations are carried out on the atomic level, the simulations by means of FEM have been done on mainly two levels: the mesolevel, where the behaviour of a set of grains representative for the whole sample is described [6,7], and the microlevel, where the grain behaviour is modeled [5,8,9].

Xu et al. [8] have carried out FE simulations of martensitic transformation in a 2D crystal embedded in a non transforming matrix. The basic transformable unit is one element of the crystal. The martensitic transformation strain is the Bain strain. Phases are considered elastic. The authors simulate the transformation process (appearance of different martensite variants) and transformation "kinetics" on the basis of a minimum free energy path.

Marketz et al. [6] and Simonsson [7] focussed on the description by FE of the behaviour of a set of grains considering 24 possible orientations in the grain (either in 2D [6] or in 3D [7]) and elastoplastic behaviour of the phases. The transformation strain is a shear in the habit plane and a volumic variation normal to the habit plane. In their transformation criterion, they consider that the plate orientation that is chosen is the one for which the driving force is larger than a threshold value. The transformation kinetics is either given a priori [6] or calculated [7].

In a previous work [5] a 2D finite element (FE) model of the grain has been developed. The plate orientations and the evolution of transformation plasticity as a function of transformed fraction have been described. But in this model the choice of the plate orientation was determined only through the maximization of the mechanical driving force associated with the transformation.

In the present work, the scale of the modelling is the same as in [5]: one plate (or variant) of martensite is considered to be made of domains. Two deformations are associated with the formation of a martensite

plate: the macroscopic shear γ_0 in the habit plane and the volumic variation ϵ_0 in the normal direction of the habit plane. The macroscopic shear γ_0 is considered as not being accommodated by the formation of new domains inside the plate, at the plate tip [10]. This scale is a "macroscopic" in comparison with modelling done at a lower scale and which considers the Bain strain as the strain to be accommodated. Thus, we assume that the substructure inside a martensite plate (twin distributions and habit plane ...) is not dependent on the stress state. This hypothesis is realistic at a first order. Moreover, we do not describe the plate growth but the way the grain is filled by successive martensite plates, and the associated mechanical behaviour. The main difference with [5] is that the transformation criterion here is more complete. It considers not only the mechanical driving force but also the chemical driving force and the strain energy increment associated with the transformation.

In the following, we describe our model. Then the results for a Cu-Zn-Al shape memory alloy and a Fe-Ni-C alloy will be analysed.

2. COMPUTER SIMULATION MODEL OF THE MARTENSITIC TRANSFORMATION

2.1 Micromechanical model

We have established a model describing the grain behaviour for a 2-dimensional geometry [11]. In a previous description, the imposed periodic boundary conditions lead to very strong constraints especially in the four corners. These constraints did not allow to describe any self-accommodating plates at the very beginning of the transformation. Thus, as Xu et al. has done [9], a medium has been introduced that surrounds a subarea where transformation will occur. The introduction of the external medium allows us to study the effect of the interactions with neighbouring grains by imposing different constraints on the transforming grain. The size of the subarea, which locates at the center of the whole mesh, is made of 14×14 elements; the whole mesh including the external medium is a grid of 22×22 triangular elements. The minimum unit that can be transformed is a band. A band is composed of elements that lie in the same direction. One band is stopped either by already transformed elements or by the grain "boundary".

The use of triangular elements allows to form 4 kinds of bands, i.e. horizontal, vertical, and 2 oblique bands. Each band can be sheared in two opposite directions and hence we have 8 different shear directions, which are 0, 45, 90, 135, 180, 225, 270, 315 degrees respectively. They can be regarded as 8 different kinds of two-dimensional variants each corresponding to one habit plane in martensitic transformation. The transformation develops by the successive transformation of bands within the grain.

To describe the configuration of martensite in a partially transformed state, we label each element by its position vector, \mathbf{r} , and define a function $F(\mathbf{r})$, which has the value 0 if the element at \mathbf{r} is untransformed and n ($\neq 0$) otherwise. Here n represents the sequence number of the transformation. All the elements transformed in the same band share the same sequence number. In the description of the final microstructure, these transformation sequence numbers and the associated shear direction are presented. In the whole process of transformation, $F(\mathbf{r})$ is kept to be 0 for the elements within the external medium. External stress is applied to the boundary of the medium. In the present paper we only consider the influence of uniaxial stresses (in x direction) upon transformation.

2.2 The phase transformation path

The chemical free energy difference between austenite and martensite during the cooling process is the driving force for martensitic transformation. The build-up of strain energy, energy dissipation, and interfacial energy are the resistive forces for it. As reported in literature [12-14], energy dissipation has an important contribution as a resistive force. To take it into account, we would need to simulate the formation of the martensite plate itself, considering firstly the formation of domains or transformation twins, and secondly the γ/α' interface mobility. This level of description is not tractable in the present simulation.

Concerning interfacial energy (restricted to γ/α' interfaces) it can be reasonably neglected as compared to the strain energy. In addition, the local stress state affects the transformation process through the work produced. Thus, the variation in free energy associated to the austenite-martensite transformation $d(\Delta G^{A-M})$ reads :

$$d(\Delta G^{A-M}) = -d(\Delta G_{ch}^{A-M}) - dW + dE$$

$d(\Delta G_{ch}^{A-M})$ is the variation in chemical free energy difference associated with the transformation of a new band of volume dV_m and is assumed as follows:

$$d(\Delta G_{ch}^{A-M}) = \Delta G_{ch}^{A-M} * dV_m = \Delta S^{A-M} * (T - T_0) * dV_m$$

T is the transformation temperature, T_0 is the equilibrium temperature, ΔS^{A-M} is the entropy variation dW is the mechanical driving force :

$$dW = \int dV_m (\sigma_n \epsilon_0 + \tau \gamma_0) dv .$$

where σ_n and τ are the normal stress to the habit plane and the shear stress in the habit plane respectively. dE is the variation of total strain energy (in both phases) due to the transformation of dV_m :

$$dE = d(1/2 \int_v \sigma_{ij} \epsilon_{ij} dv)$$

σ_{ij} and ϵ_{ij} are the components of the stress and strain tensors respectively. v is the whole volume.

The transformation of one band can occur only if $d(\Delta G^{A-M})$ is negative.

As in this paper, we only present simulation results concerning the transformation during continuous cooling under a constant applied stress, the criterion works in the following way. For the first band selection, dW and dE are computed for all candidate bands and the chemical driving force ΔG_{ch}^{A-M} is then set at the value that is just sufficient to make $d(\Delta G^{A-M})$ negative for one band. The temperature corresponding to this ΔG_{ch}^{A-M} is the so-called M_s temperature. The band with the minimum ΔG_{ch}^{A-M} requirement is chosen as the first band and is transformed immediately. Mathematically, $F(r)$ is set equal to 1 and the transformation strain is given simultaneously to all the elements in the band. To find the next band, $d(\Delta G^{A-M})$ is recomputed for all the new possible bands based on the already transformed configuration. Usually, several bands may have negative $d(\Delta G^{A-M})$ at the same temperature. In that case, the band which maximizes the decrease in free energy $d(\Delta G^{A-M})$ is chosen as the second band to transform. If no band fulfills the criterion the temperature must be decreased and this is realized by increasing ΔG_{ch}^{A-M} . This procedure gives the transformation "kinetics" i.e. the formed martensite fraction versus ΔG_{ch}^{A-M} or temperature (T) within the scale factor that relates ΔG_{ch}^{A-M} and T . The transformation path that is simulated by this procedure is a minimum energy path.

2.3 The transformation strain and the mechanical properties of the two phases and the medium

As transformation occurs, all elements of the band receive the transformation strain. In the local basis ($\underline{d}, \underline{n}$) of each band, the transformation strain tensor is written as follows:

$$\epsilon_{\underline{d}, \underline{n}}^{tr} = \begin{bmatrix} 0 & \gamma_0 / 2 \\ \gamma_0 / 2 & \epsilon_0 \end{bmatrix}$$

In this paper we do not consider plastic accommodation: austenite and martensite are assumed to behave elastically.

For Cu-Zn-Al alloy we have $\epsilon_0 = 0.003$, $\gamma_0 = 0.19$. The mechanical input data are: Young's modulus 1.10^5 MPa and Poisson's ratio 0.3 for both austenite and martensite. They are considered as temperature independent.

Concerning the mechanical behaviour of the medium, we have considered either a nontransforming medium having therefore the same Young's modulus as austenite or more generally a transforming medium (that represents the effect of surrounding grains) which has the global mechanical behaviour of the alloy. Young's modulus is taken equal to 1.10^4 MPa; this value corresponds to a measured strain of about 1% when the alloy is submitted to an external stress of 100MPa . Poisson's ratio is kept equal to 0.3.

For Fe-Ni-C alloy, we take $\epsilon_0 = 0.015$, $\gamma_0 = 0.20$. Young's modulus of austenite and martensite is 2.10^5 MPa; Poisson's ratio is 0.3. For the medium, we have chosen a Young's modulus of 2.10^4 MPa and a Poisson's ratio of 0.3 (based on the same analysis as above).

The computations are performed with the FE code SYSWELD [15] using an infinitesimal strain formalism with a plane strain assumption.

3. SIMULATION RESULTS AND DISCUSSION FOR THERMO-ELASTIC BEHAVIOUR OF Cu-Zn-Al ALLOY

Firstly, we have tested our model in the case of CuZnAl shape memory alloys. The aim is to situate our calculated results towards experimental ones found in literature.

3.1 Relation between M_s temperature and applied stress

Martensitic transformation does not take place at the temperature (T_0) of thermodynamic equilibrium between the austenite and martensite, but only after sufficient undercooling to achieve the required driving force for the transformation. It is also known since a long time that an external stress affects the transformation start temperature : Patel and Cohen [16] showed and well analyzed the approximate linear relationship between external stress and M_s temperature for iron-nickel and iron-nickel-carbon alloys. For Cu-Zn-Al shape memory alloy, we recall Patoor's results [17] in figure 1.

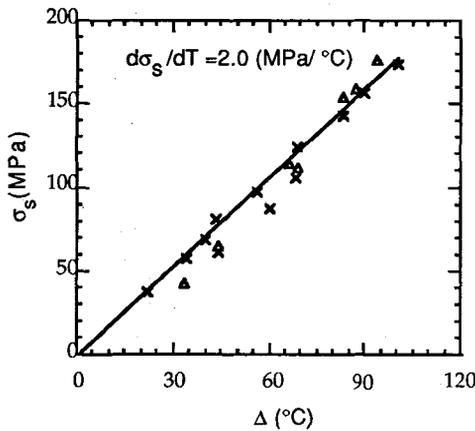


Fig. 1: Experimentally observed ΔM_s for different applied stresses [17]

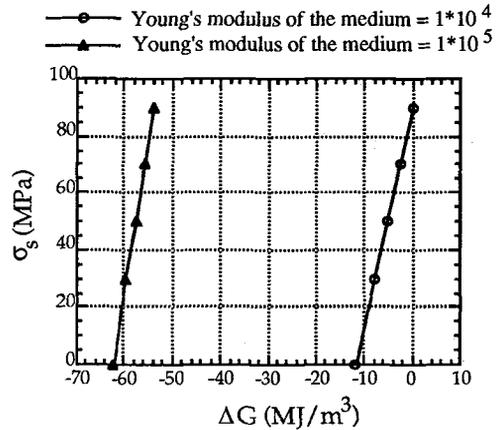


Fig. 2: Calculated ΔG_{ch}^{A-M} for different applied stresses

Patoor's experiments were carried out under either constant temperature or constant stress. A linear relationship between applied stress and temperature is obtained with $d\sigma/dT=2\text{MPa}/^\circ\text{C}$. Our calculated results are shown in Fig.2 where the minimum chemical free energy is reported for different applied stresses. In order to analyse the effect of the mechanical behaviour of the medium, results with Young's modulus equal to 1.10^5 (for the medium) are also presented on this figure. In both cases a linear relationship between the minimum chemical energy and applied stress is observed but a difference in slope appears. Considering $\Delta S = 0.23\text{MJ}/\text{m}^3/^\circ\text{C}$ [17], the calculated slopes $d\sigma/dT$ are $1.8\text{MPa}/^\circ\text{C}$ and $2.4\text{MPa}/^\circ\text{C}$ for Young's modulus equal to 1.10^4 MPa and 1.10^5 MPa respectively.

Our results show also that in the case of a rigid medium, higher undercooling is needed to make the transformation start: this is due to higher strain energies that are generated as the first plate forms. The calculated undercoolings are quite larger than the one observed experimentally. The difference can be attributed to the large amount of phase formed at the first step that is directly associated with the geometry of the mesh. As the undercooling is directly related to the strain energies and dissipation, a lower fraction of martensite formed will give a value much nearer to the experimental one.

When transformation occurs, the external medium represents the properties of the surrounding matrix being able to relax partly the stresses by transformation in the surrounding grains. For the M_s temperature, it is clear that the properties of the medium are that of the austenite; but as transformation proceeds, the surrounding grains are able to transform and thus to relax local stresses. Also, for the further calculations, the less rigid behaviour will be considered.

3.2 Transformation under different applied stresses

The influence of an external uniaxial stress on the transformation "kinetics" and on the strain evolution are presented in this part. The corresponding plate patterns are also analyzed.

3.2.1 Influence of external stress on transformation kinetics and microstructure

Fig.3 is a plot of the transformed fraction as a function of the chemical driving force under stresses that vary from 0 to 500 MPa. Three features are particularly striking from the curves. First, the transformation occurs in a burst way in the case of no applied stress. The transformation goes directly to near completion ("complete burst") once the first martensitic band is formed. Second, the transformation temperature range,

i.e. (M_s - $M_{90\%}$), ($M_{90\%}$ is the temperature at which 90% martensite is formed) increases as the applied stress increases from 0 to 500 MPa. Third, $M_{90\%}$ keeps nearly the same for all applied stresses.

To better understand these features, it is necessary to examine the microstructures that develop under different loads. Fig.4 illustrates the plate pattern of a stress-free transformation. Two variants with opposite shear (315° and 135°) directions are formed successively and have almost equal areal fractions in the final microstructure. The adjacent bands always shear in an opposite sense to form the so-called self-accommodating martensite. After one band is transformed, it is the shear component of the transformation strain that contributes a lot to the build-up of strain energy. The formation of self-accommodating martensite allows to minimize strain energy and this is the reason for the "complete burst" as observed in Fig.3. We can also notice that the transformation proceeds by transformation of adjacent bands. This behaviour can be reasonably understood if one considers on one hand that the bands with the lower width/length ratio, i.e. adjacent bands, will lead to the lowest increase in strain energies (as long as the mechanical properties of the two phases are the same) and on the other hand local stresses are the largest near the plate tips of the first plates formed and thus the additional driving force will be the largest in these areas.

As far as the comparison between experimental and calculated results in the stress free case is concerned:

- the "complete burst" is not observed: a 16°C temperature decrease is needed experimentally for a 90% amount of transformation [13]. This is equivalent to an increase of about 4 MJ/m^3 in $-\Delta G_{\text{ch}}^{\text{A-M}}$.
- although self accommodation is observed experimentally, more than two variants appear.

These differences between experiment and calculation can be related to the fact that:

- the very large amount of martensite associated with the first plate will give high internal stresses. These stresses will be relaxed by the formation of self accommodating plates. As the mechanical properties of the two phases are the same, the resistive force due to deformation incompatibilities between the different plates will not increase as the martensite content increases. Also, any modification of the mechanical properties (with the nature of the phases or with the crystal orientation (for the martensite)) will lead to an increase in the resistive force and thus limit the burst phenomenon.
- near the M_s temperature simultaneous nucleation sites can be activated leading to plates with different orientations. Consequently, larger interaction energy between plates and grain could be created and also lead to a larger transformation temperature range.
- we neglect the dissipative energy that would give an additional resistive energy for the transformation and lead to a larger transformation temperature range

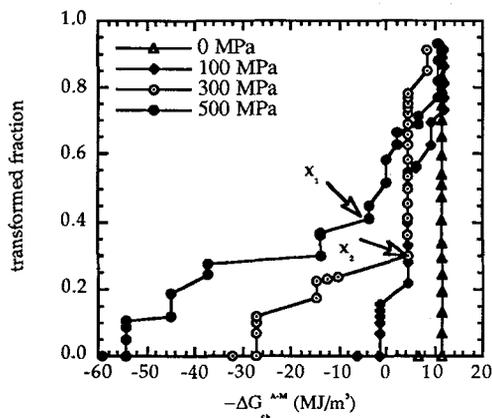


Fig. 3: Transformed fraction versus $-\Delta G_{\text{ch}}^{\text{A-M}}$ under different loads. Before x_1 and x_2 , only one variant appears

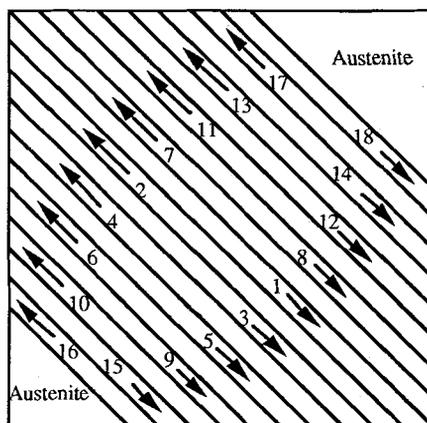


Fig.4 Stress-free transformation sequence

Under an external stress, the transformation process strongly depends on the stress level. The uniaxial stress affects the transformation in two ways. Firstly, a uniaxial tension will increase the M_s temperature (Fig. 3) as described in part 3.1. Secondly, the applied stress in x-direction is always in favour of bands sheared in 315° direction in comparison with the 135° sheared bands.

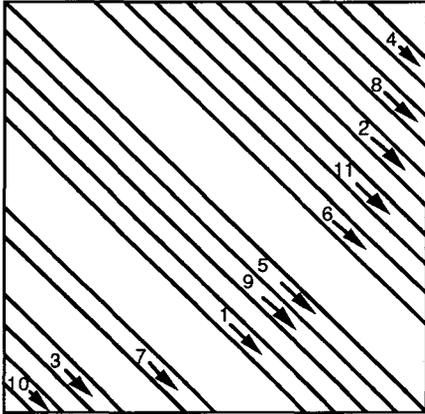


Fig.5 Transformation sequence under 500 MPa before point x_1 in Fig.3

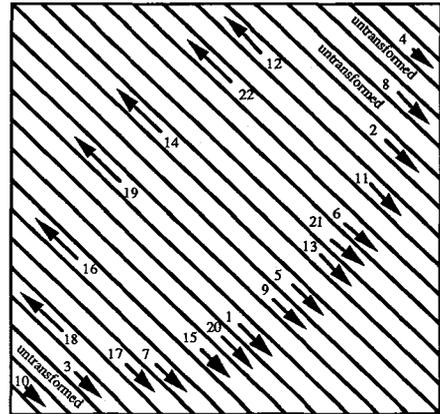


Fig.6 Transformation sequence under 500 MPa

Fig.5 shows the microstructure of a grain with about 40% martensite formed under a 500 MPa external stress (corresponding to the transformation before point “ x_1 ” in Fig.3). Only one variant (sheared in 315° direction) is present. The mechanical driving force associated with the applied stress has a predominant effect in the transformation criterion. Since the plates that could accommodate the macroscopic shear strain are not able to form, it is necessary to further cool the specimen in order to make the transformation progress. This explains the spread in the transformation temperature range (Fig.3). Self accommodating plates are observed above 40% martensite formed (Fig.6) at a temperature slightly above the M_s temperature (without applied stress). The further transformation occurs like a burst (Fig.3) as in the case of no applied stress.

The cases with lower applied stresses behave quite similarly. However, as the external stress decreases, the orientation effect decreases : the amount of martensite with a favourable orientation towards the applied stress decreases. Besides the fact that M_s decreases, it is also noticeable that $M_{90\%}$ temperature keeps almost the same irrespective of the magnitude of the external stress.

3.2.2 Macroscopic strain evolution under different loads

The macroscopic strain is defined as the total strain of the cell (grain+medium). Here we show how the applied stress affects the macroscopic strain.

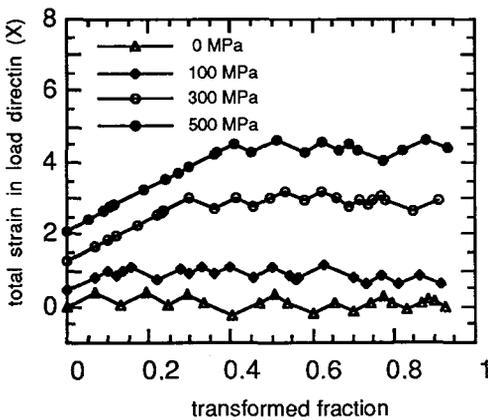


Fig. 7: Macroscopic strain versus transformed fraction

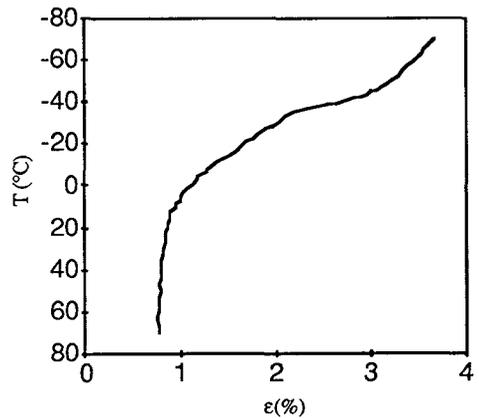


Fig. 8: Experimentally observed deformation against temperature under 116 MPa external stress [17]

Fig.7 is a plot of the macroscopic strain versus transformed fraction for different loading cases. At the earlier stage of the transformation, a linear increase of strain is observed due to the favourable plate orientation. When the transformation comes to the point x_1 or x_2 (Fig.3), as already described, the orientation effect decreases. Consequently the strain becomes nearly constant. (Two typical slopes appear on the curves that correspond to the transformation of 315° and 135° variants respectively). With our elastic assumption, we show that it is mainly the orientation of the martensite plates that controls the deformation level of the alloy. This is in agreement with experimental knowledge.

It is interesting to compare the calculated strain variation versus the increase in chemical driving force to the experimental strain measurement versus temperature (Fig.8 [17]).

In order to test our model, we have carried out a simulation that follows this experimental condition.

The result is shown in Fig.9. At each recorded $(-\Delta G_{ch}^{A-M})$, there are always several bands that can be transformed. The strain corresponding to the last band is recorded as the strain under this ΔG_{ch}^{A-M} . The simulated result shows the same tendency of variation of the strain versus temperature. However, we see that the total strain is much smaller than the experimental values due certainly to an overestimation of the self accommodation.

4. SIMULATION RESULTS AND DISCUSSION FOR Fe-Ni-C ALLOY

The transformation in Fe-Ni-C alloys is characterized by a large volume change and in most cases the occurrence of plastic accommodation [2]. Nevertheless, under our assumption of elastic behaviour, we can compare the behaviour of the two alloys.

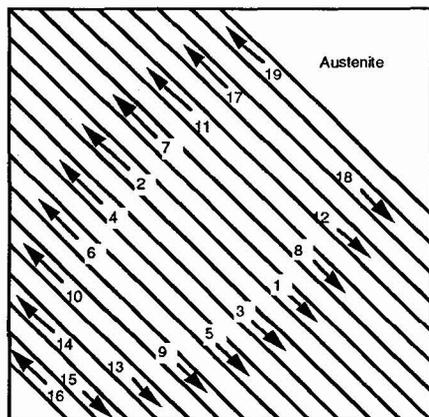


Fig.10: Stress-free transformation sequence

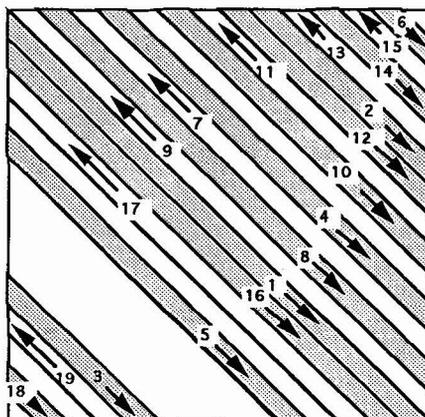


Fig.11: Transformation sequence under 500 MPa

Let us first compare the results concerning the transformation sequence (Fig. 10 and 11), transformation "kinetics" (Fig. 12) and global deformation of the cell (Fig.13) in the present Fe-Ni-C alloy with those obtained in the Cu-Zn-Al alloy. The difference in behaviour between the two alloys is related to the difference in local stress level due to the different input data. For the Fe-Ni-C alloy, a higher Young's modulus and a higher volume change are taken into account. These will definitely lead to a higher local stress level.

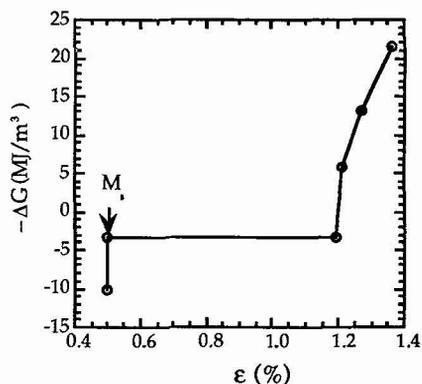


Fig. 9: Calculated total strain vs $-\Delta G_{ch}^{A-M}$

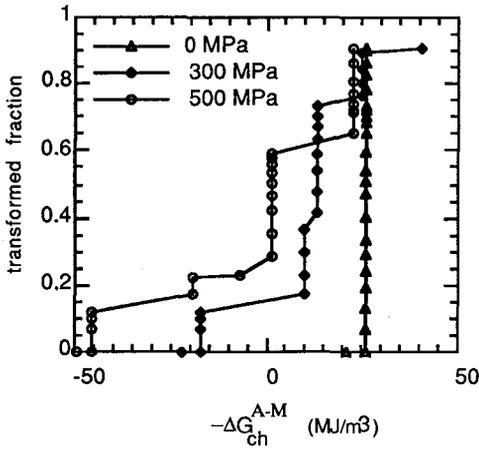


Fig. 12: Transformed fraction versus $-\Delta G_{ch}^{A-M}$ under different loads.

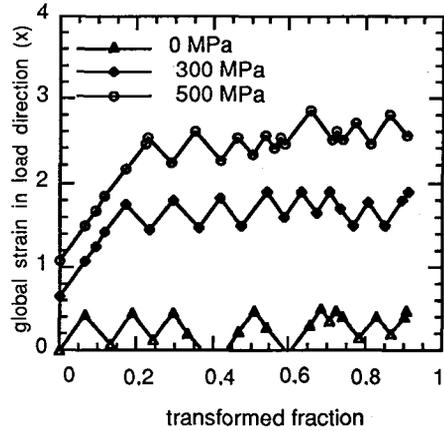


Fig.13 Global deformation versus transformed fraction

The stress-free transformation sequence (Fig.10) shows a fully self-accommodated plate pattern as in the case of Cu-Zn-Al alloy. We note that the detailed sequence is not exactly the same due to differences in local stress states. As for CuZnAl, a complete burst transformation occurs as shown on the transformation “kinetics”(Fig.12). But, for the FeNiC alloy it comes out that a larger chemical energy contribution is necessary to start the transformation due to larger strain energies (higher local stresses).

The influence of a 500 MPa external stress on the transformation sequence is shown in Fig.11. The 135° variant appears after six 315° variants (favourably oriented variants by the applied stress) are formed, i.e. after about 23% transformed fraction. For the Cu-Zn-Al alloy, this fraction was about 41% (Fig.5). This result shows that the effect of the external stress is much weaker in the present case than in that of Cu-Zn-Al alloy due to the higher internal stress level in the former alloy. Consequently, the final deformation (Fig. 13) remains at a lower level due to a smaller contribution of the orientation effect.

To well distinguish the effect of the two sources of higher internal stress, i.e. higher Young's modulus and volume change, a simulation with a pure shear transformation strain has been performed. For a stress free transformation the results show exactly the same sequence as in Figure 10 up to the 8th band which represents a 46% transformed fraction. It seems that the volume change part of the transformation strain does not play a significant role at the earlier stage of the transformation. This means that the transformation is mainly controlled by the shear component at that stage. However, as the transformation goes on, some differences appear in the transformation sequence (different positions of the successive bands). They can be related to the fact that the stress state is different with the volumic change and thus the transformation sequence is slightly modified. This analysis can be extended to the case of an applied stress: the same sequence as in figure 11 is found up to the 17th band (about 71% transformed fraction). Afterwards, differences in the positions of the self accommodating bands appear. It must also be noted that the transformation “kinetics” and the global deformation are quite similar for the pure shear transformation and for the transformation with a volume change.

Finally, we concluded that for the present simulations, the difference of Young's modulus between the two alloys is more effective on the local stress states than the difference of the volumic variation associated with the transformation.

5. CONCLUDING REMARKS

The martensitic transformation in both Cu-Zn-Al and Fe-Ni-C alloys is simulated in the present study. The transformation occurs in a grain that is embedded in an surrounding medium. The results in Cu-Zn-Al shape memory alloy illustrate the influence of the applied stress on the mechanical behaviour, the transformation kinetics and the microstructure (plate pattern). For the transformation kinetics, three features are particularly striking. i) The stress free transformation is conducted in a burst way; the transformation strain is accommodated by the formation of self-accommodating martensites. ii) The transformation temperature range increases as the load is increased. iii) $M_{90\%}$ keeps nearly the same for all loads tested.

The results reveal the general tendencies of M_s variations, plate patterns, and associated mechanical behaviour for the transformation under stress. However, discrepancies remain that are related to the assumptions considered in the modelling.

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