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Experimental Studies, Modelling and Simulation of the Hysteresis in SMA Single Crystals: The σ, ε, T and t Coordinate Space

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Abstract: The behaviour of global and partial loops (stress-strain-temperature) in SMA single crystals was investigated. Marquardt's based numeric algorithms were developed for the identification of relevant physical parameters (time independent). In addition, the time-dependent effects like static and dynamic recoverable creep, β-recovery, quenching and or up-quenching effects, were evaluated. The constructed model permits the computer simulation of the global and partial hysteresis cycles in the four dimensional σ, ε, T and t coordinate space. The simulated curves are in excellent agreement with the experimental results.

1. INTRODUCTION

SMA are one of the so called smart materials, because of the shape memory effect (one way and two-way), the pseudoelasticity effect, and its applications. The applications require a rigorous stability of transformation temperatures and a small hysteresis. The design and construction of SMA devices can be greatly improved if models, describing properly the behaviour of the working point of the material in the stress-strain temperature (σ, ε, T) coordinates, are available. These models (which are time independent and free of the irreversible actions of the external fields) are the basis to include the experimental time effects: martensite "creep", stress evolution at constant strain and the micromemory.
In this work, the physical parameters used in the model (assembly of single variant martensite plates) are estimated. From experimental curves (hysteresis cycles, partial loops and the temperature dependence) an identification routine is established. They provide a physical image of the material. In the experimental analysis, particular attention to the occurrence of diffusional processes and the consequent dependence of the hysteretic behaviour with time was paid. Different behaviours were identified, associated to the diffusional processes (stabilization activated by the presence of interfaces and related to the martensite "creep", and β-recovery). A time-dependent model was put forward based on the observed elementary processes.

2. EXPERIMENTAL

Two experimental high resolution stress - strain - temperature equipment (see, for instance, [1]) have been used. Sometimes, thermomicroscopy and resistance measurements were used. The cycles were produced at low stress-rates ($d\sigma/dt < 1$ kPa s^{-1}) because at higher values ($d\sigma/dt > 10$ kPa s^{-1}) a widening of the hysteresis cycle is observed, associated to changes of the specimen temperature due to the release of latent

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heat. A Cu - 14.6 at% Zn - 16.7 at% Al high purity single crystal was used. The nominal M_S temperature was 288 K. A planar specimen was cut with a low speed saw and a neck shape was produced with a fine blade. The sample was subjected to a solution treatment at 1120 K for 30 minutes followed by quenching in cold water (TT2). Eventually, air quenching (TT1) is used. After the thermal treatment, the specimen was carefully polished. The cross section of the sample was ≈ 1 mm². From the initial water quench, about 1000 transformation cycles were done in a period of near 1 year. Afterwards an "up-quenching" (3 days at 340 K) was done, followed by air cooling to the working temperature of ≈ 290 K. In all the experiments only a small central portion of the sample (near 10 %) was transformed.

3. TIME INDEPENDENT BEHAVIOUR

From the experimental observations it was found that the behaviour of a single martensite plate can be schematized, in the f-x coordinates, as shown in figure 1a. The ABCD-DCBA path represents the reversible behaviour and the AB'B"D-DC'C"A path the real behaviour, including nucleation of both phases (martensite as also austenite). Such behaviour can be well represented by the mechanical system (derived from [2]) depicted in figure 1b, which uses a spring with joined coils. This last condition explains the temperature dependence of the transformation stress. If the external temperature increases, the stress joining the coils of the spring increases as an action of the Clausius-Clapeyron equation. The supplementary stress to overcome the temperature effect equals $(df/dT)_{CC}(T_{ext} - T_0)$. The different parameters associated to a single plate are schematized also in figure 1b. It should be noted that a mechanical spring model (that can be considered as derived from ref. [2]) has been also used to describe the hysteretic behavior in polycristals [3]. However, in such model the parameters remain only phenomenological and there are not related to the actual physical events existing in the polycristals, which are rather more complex than in single crystals.

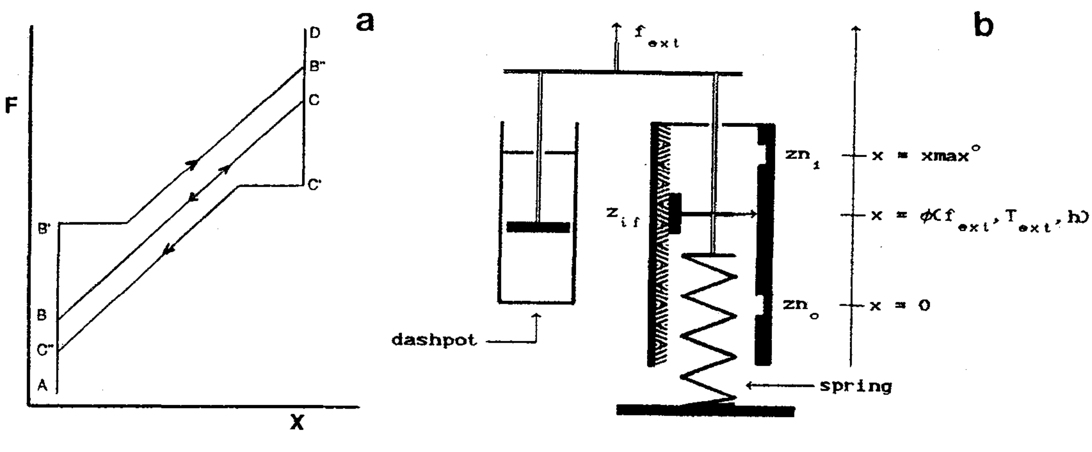

Figure 1: a) Schematic behaviour of stress against strain (force F versus elongation x) for a "single" martensitic plate; ABCD-DCBA reversible path. b) Mechanical analogy: z_{if} interface friction; zn_1 and zn_0 nucleation; f_{ext} and T_{ext} external fields; h previous history; $xmax^0$ maximum elongation due to the plate.

Initial parameters [1] are determined from an experimental curve (figure 2). They allow the representation of the hysteretic behaviour of a single crystal in the f, x and T space, by generalizing the model to N independent plates (experimental values of f are noted F). It can be defined:
- The plate number N (usually chosen to be ≤ 100 for easy of calculation).

- The maximum macroscopic elongation Xmax which is directly deduced from the complete transformation curve (see figure 2a). The elongation associated to the each plate $xmax^0$ is defined as $xmax^0 = Xmax / N$.
- The interface friction z_{il} is linked to the displacement of the interfaces. Their values are determined from the internal loops by $z_{il} \approx \Delta F_h$ (see ΔF_h in figure 2a).
- The critical force f_{00}, which in the absence of frictional processes corresponds to the appearance of the first domain of martensite (see f_{00} in figure 2a).
- The intrinsic pseudoelastic effect $(df/dx)_0$. It is obtained as $(df/dx)_0 \approx \alpha N$; where $\alpha = \Delta f/\Delta x$ (the average slope).
- The f_{rate} represents the progressive appearance of plates (critical force difference between consecutive plates). The f_{rate} value can be obtained from the following approximate relationship:

$$\Delta f_T \approx N\, f_{rate} + \alpha\, Xmax$$

- The nucleation of martensite and of β (zn_0 and zn_1) are taken as:

$$zn_0 \approx zn_1 \approx \alpha\, Xmax$$

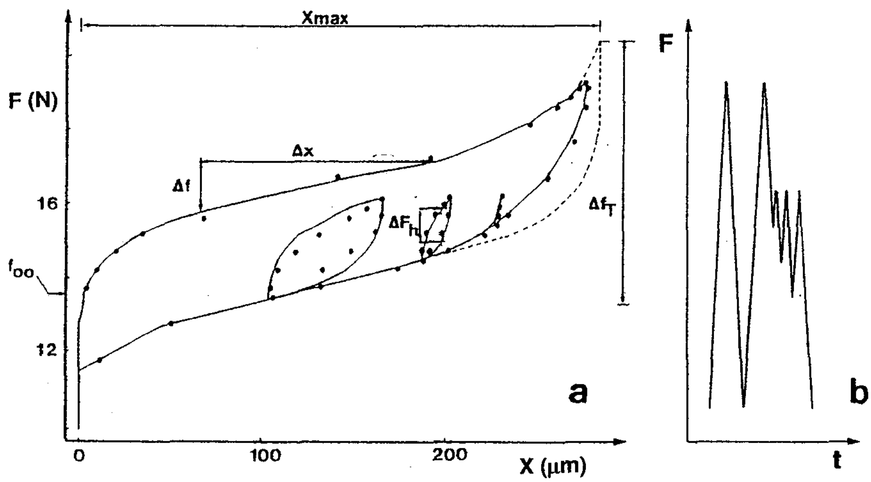

Figure 2: a) Force (F) against elongation (x) for the first approach to the physical parameters (— experimental curve, ···· simulation, ---- construction of the complete transformation cycle). b) Force against time (schematic).

The initial value of each parameter is then adjusted using a technique based in the Marquardt algorithm [5]. It is assumed that there is a functional relationship between the variables (f, x, T or σ, ε, T) of the system, through the parameters of the model and it is based on the minimization of an error criterion by iteration. Attention was paid to the instabilities produced by the frictional contributions. The simulation presented in figure 2 shows the adequate prediction of the transformation cycle obtained by the model. The parameter so obtained are the basis for the time-dependent analysis.

4. TIME DEPENDENT EFFECTS AND MODELIZATION

One of the aspects to be taken into account is the M_s temperature evolution after a quenching and also after a "micro up-quenching". Using an initially air-quenched Cu-Zn-Al single cristal, a treatment of 10 hours at 348 K produces an increase of the critical transformation force f_{00} of near 1.40 N. It then diminishes with time (figure 3). In this particular case a change in M_s (or T_0) of 1.71 K is found.

In order to separate the different contributions, associated to the coexistence between the phases, further experiments were done far enough from the initial thermal treatment of a water-quenched Cu-Zn-Al single cristal [6]. The following step was to obtain a quantitative approach to the "martensite creep" (growth of martensite with time at constant stress and temperature in a partially transformed state). Experiments are carried out arriving to the initial point at a stress rate near zero ($\leq$ 1 kPa s^{-1}). This type of phenomenon is called "Static Recoverable Creep" (SRC).

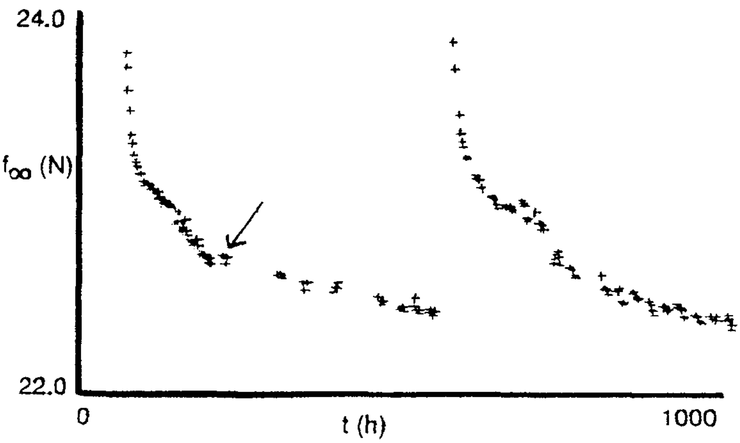

Figure 3: Critical load (f_{∞}) against time (t) after two similar "micro up-quenching". The arrow shows the influence of the smooth cycling on the evolution (from 12 to ≈ 3 cycles/day).

In figure 4a the hysteresis cycle with a typical SRC experiment is shown, and in figure 4b the corresponding elongation against time. The hysteresis cycle is locally deformed in the retransformation (see the arrow in figure 4a). This is classically known as micromemory. The normal shape of the hysteresis cycle is recovered after a few minutes in parent phase, but not instantaneously (a "β-recovery" process). The maximum deformation is, within the experimental scatter, the same in each point of the linear part of the transformation curve and lower outside. It is possible to associate this phenomenon with the coexistence of both phases (or with the presence of interfaces).

In the language of the model, the SRC produces the growth of active plates (plates with β and martensite coexisting) and leaves unaffected areas which are completely in β or in martensite. The SRC effect during the unloading part of the hysteresis cycle is less marked and a minor initial reduction of length can be detected, probably associated with the recovery of β in the neighbouring of the retransformed parts. The fact that the barrier (interface friction) producing the hysteresis plays against the growth of the martensite, explains the reduced activity of the SRC on unloading.

An increase in complexity is achieved by loading the material at relatively higher stress rates (1 kPa s^{-1} $\leq$ dσ/dt $\leq$ 10 kPa s^{-1}), which is called "Dynamic Recoverable Creep" or DRC. The behaviour is similar to SRC but more pronounced, as shows the DRC elongation against time in figure 4b.

In the particular case of periodic oscillations an important evolution exists as it can be seen in figure 5a. The results indicate that the zone of the material affected by the cycling (related to the range of forces used) tends rapidly to the martensitic state and eventually produce the appearance of neighbouring plates. On unloading, a more important stabilization (or micromemory) is observed (see the arrow in figure 5a). The macroscopic actions in parent to martensite are connected with local stabilization (increase of T_0) processes activated by the existence of an interface and its movement. In retransforming to parent phase the main action is associated to the β-recovery process (decrease of T_0).

As a consequence of previous results, some time-dependent contributions are added to the time-independent model [7] in the form of changes of the equilibrium temperature T_0 of the material [6]. The computer program considers the material as formed by β domains which can transform partially or totally to martensite [7]. The time-independent parameters were obtained from [7,8]. The action of time over the system configuration is taken into account as an appropriate shift of the equilibrium temperature of each domain. In the special case of the DRC actions, a further interaction with near-neighbouring domains is considered. From the equilibrium temperature, the action of the external fields (experimental force F_i and temperature T_i) is calculated for each time t_i using the Clausius-Clapeyron equation. The result gives the elongation associated to the each domain $x_i(j)$, $(1 \le j \le N)$ and consequently the total increased of length is $x_i = \sum_{j=1}^{N} x_i(j)$. The next iteration follows from the analysis of the new equilibrium temperatures.

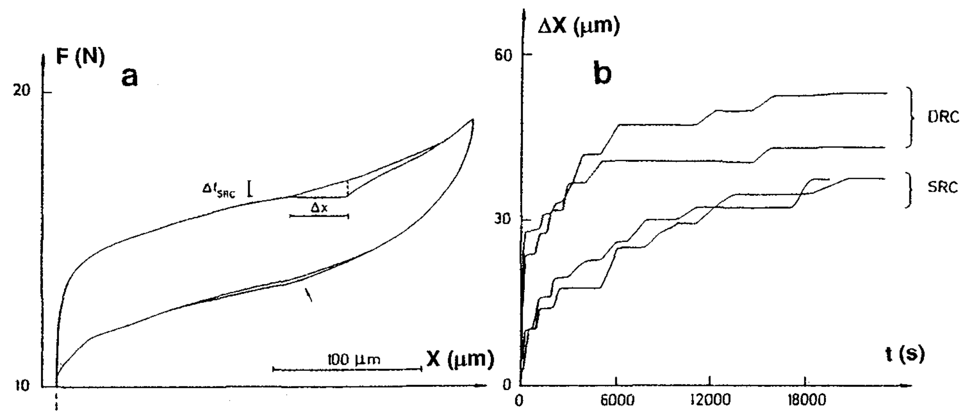

Figure 4: a) Force (F) against elongation (x) and time effects; Δx is the creep elongation and Δf_{SRC} the load change related to the local equilibrium temperature variation. The arrow indicates the slight stabilization. b) Creep (Δx) against time (t) during SRC and DRC.

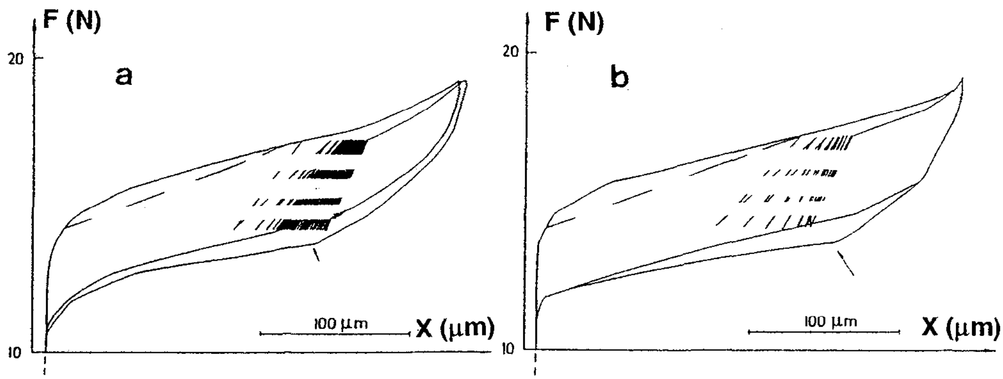

Figure 5: Force (F) against elongation (x), internal cycling (only extreme points are measured) and time effects. a) Experimental curve: stabilization or micromemory (see the arrow) is accentuated by the DRC cycling effect. b) Simulated curve using the f, T, t experimental data.

In the general case, the whole complex system will be simulated as a sum of the simpler SRC and, if appropriate, DRC cases. The complete description of these cyclic processes must take into account the growth, shrinkage and eventually, the nucleation and disappearance of plates, together with frictional and quenching effects (see figure 5b).
Most of the observed experimental results (if one avoids the creation of dislocations) can be explained with this model. For example, the evolution with time of T_0 after a heat treatment explains easily the appearance of an "isothermal" transformation and also the difference between hysteresis cycles belonging to two distinct moments. Micromemory (independent of dislocations creation) can be associated to the local recoverable equilibrium temperature changes.

5. CONCLUSION

From high resolution experiments, some time-independent physical parameters have been found, which allow to modelize SMA thermomechanical behaviour. The observed time effects are linked with the stabilization activated by the interfaces, the recovery of the β phase and the previous quenching treatments. They can be explained as a recoverable change of the equilibrium temperature T_0 of each plate. It allows to interpret and modelize martensitic recoverable creep effects (increase of T_0) and micromemory (local change of T_0) as actions due to the change of atomic order accelerated by the coexistence of the phases.

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