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Propagation of an austenite–martensite interface in a thermal gradient

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Abstract. The mechanism of thermally driven shape recovery from a single variant of 2H-martensite to the parent austenitic phase is experimentally studied on a specimen of Cu–Al–Ni shape memory alloy (SMA). The formation and motion of the martensite-to-austenite transient interfaces is controlled by a thermal gradient, and recorded by a CCD camera. Independently, the moving boundaries are observed by an infrared camera to capture the temperature evolution accompanying the propagation. Both the velocity profiles of the propagation and the thermal images indicate that the shape recovery of SMAs is a complex dynamic mechanism, which cannot be described by a classical Stefan's model of phase transitions, known from the thermal conductivity problem.

Key words: shape memory alloy, martensitic transition, phase boundary, Stefan's problem.

1. INTRODUCTION

In the past two decades, unique thermomechanical properties of shape memory alloys (SMAs) became a subject of intensive theoretical investigation. Equilibrium formation of martensitic microstructure and interfacial structure between austenite and martensite is described by Ball and James [1] by means of minimization of multi-well free energies. Similarly interesting topics have appeared in theoretical description of the martensitic transition itself, i.e. in the dynamics of propagating austenite-to-martensite (or vice versa) interfaces and twin boundaries. A concept of representing the interfaces by solitary waves (see [2] for main ideas and further references) was investigated in the 1990s, but thermodynamic models of discontinuity front propagation [3,4] became the most widely used theoretical approach. This theory has been verified for rapid, shock-induced motion, as the austenite-to-martensite transition experimentally studied by Clifton and Escobar [5], where the velocities of moving interfaces are in the order of 10^1 m s^{-1}. However, the stress-induced austenite-to-martensite boundaries (including the shock-induced transitions investigated in [5]) can have a complicated structure, combining the planar austenite-to-2H-martensite boundary with systems of thin needles of the tetragonal 18R-martensite [6]. Thermally driven boundaries in a stress-free state described in this paper are purely planar, and move as slow as 10^{-3} m s^{-1}. In real applications, where the thermal effects are combined with applied stresses, one can expect the resulting velocities to cover the whole range from 10^{-3} m s^{-1} to 10^1 m s^{-1}.

The main aim of this paper is to describe the observed thermally activated shape recovery of SMAs, and to give an experimental evidence that the martensite-to-austenite boundary can, in stress-free conditions, propagate in a wide range of temperatures and thermal gradients. The only use of the thermal gradient for driving the martensitic transition in SMAs is documented in the work of Salzbrenner and Cohen [7], who studied the thermal hysteresis of Cu–Al–Ni single crystals. However, neither the velocity profiles nor the morphology of the moving boundaries are presented in [7].

2. THEORY

The theoretical description of thermomechanical properties of SMAs originates from microstructural mathematical models of martensite. As it can be found in the widely cited book by Bhattacharya [8], in extensive theoretical work by Ball et al. [1,9,10], and many other publications, the nonlinear elasticity model based on free energy with multiple minima can be used for reliable description of a quasi-static and isothermal microstructure formation. When the boundary is set in motion, the isothermal condition, as well as the assumption of quasi-statics of the microstructure formation, is no more accessible, as the propagating boundary generates (or absorbs) latent heat and generates heat by dissipation. A common approach to this problem is to consider the boundary as a moving surface of discontinuity in the deformation gradient, regardless of how the fine structure looks like close to the interface [3,4]. Involving the microstructure in such dynamic models appears to be extremely complicated.

In our case of a thermally induced transition, another possible approach is to consider the motion of the boundary as a solution of a classical Stefan's problem, i.e. a problem of a phase boundary propagation between two heat-conducting media [11]. This model requires the boundary to have a fixed transition temperature, which was experimentally verified for gentle thermal gradients in [7]. However, our observation of the moving boundary in the infrared domain (section 4) contradicts such assumption.

3. EXPERIMENTAL METHODOLOGY

A shape recovery mechanism of a Cu–Al–Ni single crystal driven by a thermal gradient was studied. The gradient, instead of homogeneous heating, was expected to prefer the boundary propagation in a given direction and to enable observation of the motion of the boundary in a wider range of temperatures.

The specimen used for the experiments was a prism $(12\,\mathrm{mm}\times\ 3\,\mathrm{mm}\times 3\,\mathrm{mm})$ cut from a single crystal of Cu–Al–Ni alloy, with mass density of 7.055 g/cm^3 (the same value for both the austenitic and the martensitic phase). The experimental procedure was performed as outlined in Fig. 1a. The austenitic specimen was pressed in its axial direction, which induced a transition into a single variant of the 2H-martensite. To minimize the irreproducibility of the experiments due to the nucleation effects, a small nucleus of the austenitic phase was initiated in one corner of the specimen by rapid localized heating, using a gas burner.

Then, the specimen was placed between a copper heater and a cooler with temperatures driven by a pair of Peltier cells, and recorded by thermometers (see Fig. 1b for the experimental arrangement) such that the nucleus of the austenitic phase was situated on the heated side. Thus, the boundary was expected to propagate from this nucleus towards the cooler. The thermal contact between the cooling/heating device and the specimen was provided by a heat-conducting grease. To insure an as stress-free state as possible, the specimen was placed in a pair of thin aluminium plate cradles, which protected the specimen from falling from the

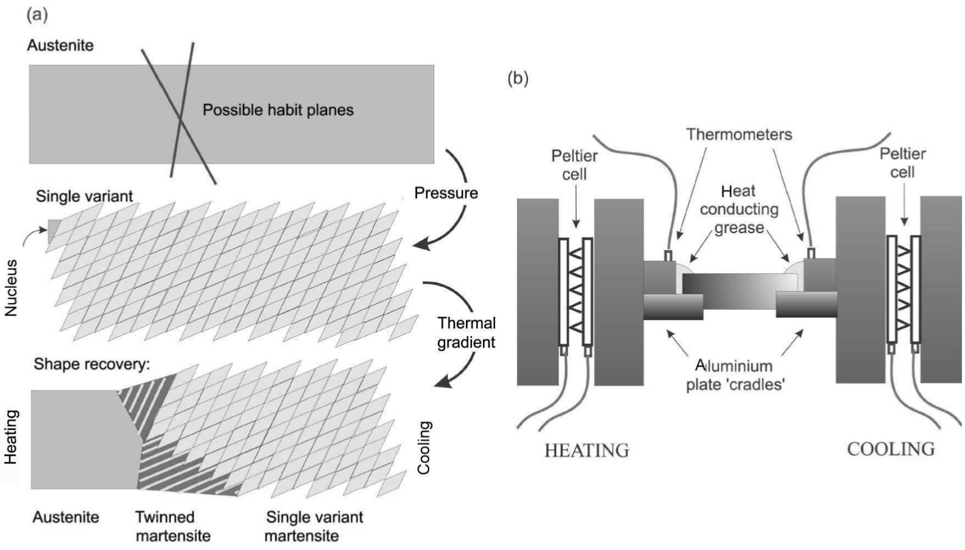

Fig. 1. (a) Outline of the experimental procedure. (b) Experimental setup for the observation of thermally induced, stress-free shape recovery of a SMA specimen.

Table 1. Orientations and dimensions of rectangular faces of the specimen. Direction angles denote deviation of outer normals from principal axes of the corresponding phases

Faces	Orientation in austenite, deg	Dimensions in austenite, mm	Orientation in martensite, deg	Dimensions in martensite, mm
A, C	[12.1; 102.1; 88.9]	11.85 × 3.18	[58.1; 88.8; 31.9]	11.01 × 3.30
B, D	[78.3; 11.7; 89.8]	11.85 × 3.13	[34.3; 89.8; 124.3]	11.01 × 3.23

device, but put nearly no constraints on its bending and torsional motion. For the same reason, the frictionless motion of the heater in the axial direction was enabled.

The crystallographic orientation of the specimen in both phases as well as the change in the geometry after undergoing the stress-induced transition into a single variant of the 2H-martensite can be found in Table 1.

The thermal gradient on a specimen was slowly steepened by increasing the temperature of the heater, T_{max}, which varied between 45 °C and 95 °C, whereas the temperature of the cooler, T_{min}, was held approximatively between 5 °C and 25 °C. After the gradient had reached a critical value, the boundary started moving through the specimen. The critical values of the thermal gradient are plotted in Fig. 2a versus the temperature of the heater, T_{max}. The critical gradients and temperatures are naturally dependent on the shape and size of the nucleus, but, in general, the interface was observed propagating in a wide range of temperatures.

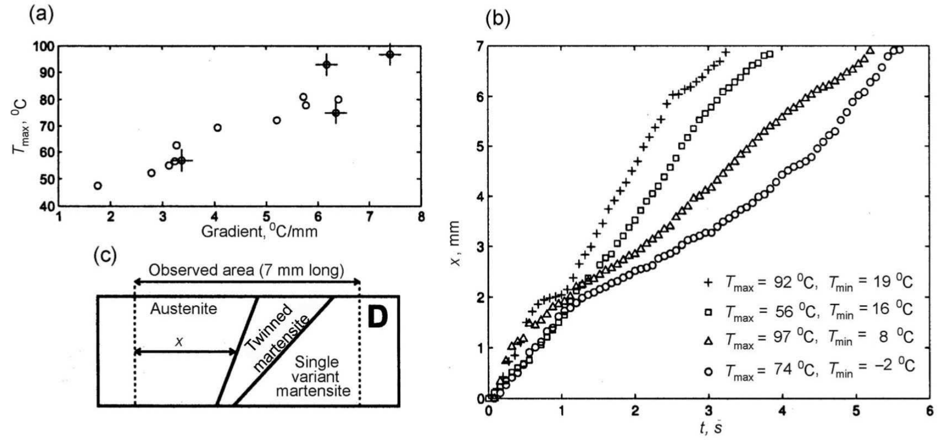

Fig. 2. (a) Plot of heater temperatures (T_{max}) versus critical thermal gradients. Crosses denote the experiments analysed in Fig. 2b. (b) Plots of interface position versus time of propagation for two different thermal gradients. (c) The position x was measured from the edge of the observed area to the middle of the observed face. Capital D denotes the observed face by notation introduced in Table 1 and outlined in Fig. 3.

The motion was recorded by a CCD camera in full PAL (640 × 480 pts) resolution. The resulting video-files were then analysed by common image processing tools of MATLAB to obtain a record of velocity of the austenite-to-martensite and twinned-to-detwinned interfaces. As shown in Fig. 2b, the velocity is not constant in the course of propagation. This figure shows four examples of position vs. time profiles in four different thermal gradients, recorded in a central section of the specimen. The position x stands for the distance from the edge of the observed region to the middle of the moving austenite-to-martensite interface (Fig. 2c), and $t = 0$ is identified with time when the interface enters the observed region. Depending on the thermal gradient, the interface can either accelerate or decelerate, but, in general, the entire microstructure always propagates together as one object. This fact will be discussed in greater detail in the next section.

4. MORPHOLOGY AND THERMAL FIELD OF THE MOVING INTERFACE

As emphasized in the theoretical section, the connection between a single variant of martensite and the parent austenitic phase is provided by a twinned martensitic structure, which is able to satisfy the compatibility conditions both with the austenite (over the habit plane) and with the single variant. As a result, the propagating interface adopts the shape presented in Fig. 3. The transition front is a combination of two independent habit planes. The intersection of these habit planes creates a coexistence line of the austenite with the single variant of martensite, whereas the regions ahead of the habit planes are filled by a twinned structure, as the compatibility conditions require. This X-like shape of the boundary is similar to the one observed by Shield [12], and theoretically derived by Hane [13], during stress-induced transition in Cu–Al–Ni single crystals working in a superelastic regime. However, Shield concludes that the two intersecting boundaries he observed can be considered as kinematically uncoupled. This conclusion cannot be valid for our case of a thermally driven boundary, which, obviously, reaches and retains one

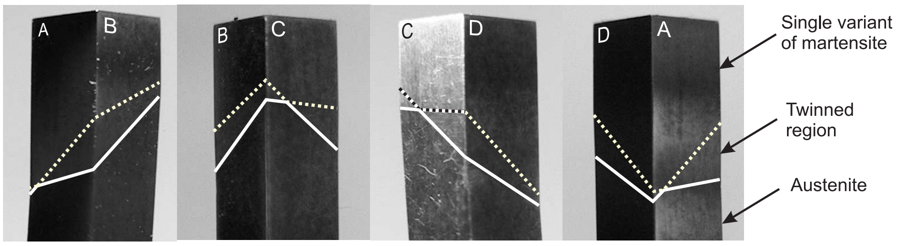

Fig. 3. Macroscopic morphology of the interface. Capitals A, B, C, D denote particular faces of the specimen, as introduced in Table 1. Dashed lines separate the single variant of martensite from twinned structures, solid lines denote boundaries between twinned martensite and austenite.

shape during most of the propagation. The stability of the shape was studied by observing the evolution of the interface from variously shaped nucleuses, or from kinked or otherwise distorted interfaces. Before starting the stable propagation, the boundary always reached the X-like shape.

To investigate the temperature field evolution during the propagation, the heated end of the specimen (including a nucleus) was recorded by an infrared camera. For these experiments, side C of the specimen was painted with a mat black colour. Significant rapid changes of the thermal field were observed at the start of the propagation. Figure 4 presents an illustrative example of such thermal field evolution. Here, the time $t = 0$ corresponds to the first observable change of the thermal field around the nucleus. The boundary starts to propagate in a thermal gradient between $T_{\text{max}} = 57\,^{\circ}\mathrm{C}$ and $T_{\text{min}} = 21\,^{\circ}\mathrm{C}$ by a rapid and strongly localized cooling ($t = 160$ ms, the second image in Fig. 4), as the forming interface moves faster than the corresponding thermal waves. Then, the propagation continues through the cooled specimen ($t = 600$ ms, the third image in Fig. 4) at lower velocities. The thermal field changes slowly until a linear gradient is reached again. Such dramatic changes in temperature cannot be observed around a stable, slowly propagating interface, where all the heat absorbed by the transition is immediately saturated by conduction, and the only observable thermal effect is a negligible global cooling of the specimen. Although at $t = -400$ ms the nucleus is stable at a temperature higher than $50\,^{\circ}\mathrm{C}$, the fully formed interface propagates through areas of significantly lower temperatures. Thus, the temperature of the interface varies during the propagation.

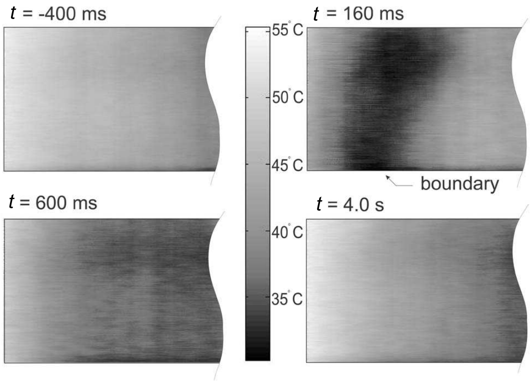

Fig. 4. The infrared image sequence of the heated end of the specimen during phase transition. The displayed temperature scale corresponds to emitivity of polished copper (material of the heater). The observed face – C. $t = -400$ ms: constant thermal gradient 400 ms before the start of the propagation; $t = 160$ ms: rapid localized cooling around the forming interface; $t = 600$ ms: specimen completely cooled through by the moving interface; $t = 4$ s: thermal gradient restored.

5. CONCLUDING REMARKS

This paper provides experimental evidence of a slow, thermally induced propagation of an austenite-to-martensite interface. As can be concluded from velocity profiles in Fig. 2b, the interface can both accelerate and decelerate, depending on the thermal gradient. This indicates that the observed motion does not obey a classical Stefan's model, where the position of the boundary is proportional to $\sqrt{t}$. Observation with an infrared camera shows that the temperature on the interface is not constant. This fact is one of the reasons for concluded discrepancy between our observations and the classical Stefan's model. More complex models, including the microstructure formation on the interface, are desired.

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Austeniit-martensiitpiirpinna levimine soojusgradiendi toimel

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On uuritud esialgse kuju soojuse mõjul taastumise mehhanismi 2H-martensiitsest vormist austeniitsesse algfaasi Cu–Al–Ni kujumäluga sulamist katsekehas. Martensiit-austeniitpiirpinna tekkimist ja liikumist on suunatud soojusgradiendi abil ja fikseeritud CCD-kaameraga. Piirpindade liikumisega kaasnevaid temperatuurimuutusi on jälgitud ja registreeritud infrapunakaameraga. Nii liikumise kiiruste profiil kui ka soojuskujutis näitavad, et kujumäluga sulamite (KMS) kuju taastumist iseloomustab keeruline dünaamiline mehhanism, mis ei ole kirjeldatav klassikalise soojusjuhtivuse ülesandest tuntud faasiüleminekute Stefani mudeliga.