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SYSTEMATIC STUDY OF THE MARTENSITIC TRANSFORMATION IN A Cu–Zn–Al ALLOY. OPTICAL MICROSCOPY AND SIMULTANEOUS THERMOSONIMETRY OF A MICROPLATE

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ABSTRACT

Optical observations, simultaneous with the detection of acoustic emission during the martensitic transformation of a 68.5Cu–14.9Zu–16.6Al (at%) show that: (a) the recorded acoustic emission associated with a martensite microplate is reproducible, and for each event the activity and the echoes last about 1 ms; (b) the intensity of acoustic emission is more important in the reverse transformation; (c) the acoustic activity is related to accelerations in the movement of the martensite microplates. The results for the observed microplates indicate the following. (1) During the process of forward/reverse transformation two kinds of acoustic emission are detected, only one of them being associated with the appearance/disappearance of visible martensite domains in the optical microscope (dimensions about 1 μm), while the other, equivalent to the acoustic emission of the β phase, and with comparable intensity to that produced by a microplate, is not related with any observable microplate, and is probably connected to lesser changes in the surface martensite. (2) The acoustic emission peaks can be related to the existence of pinnings, created by defects, in the martensite domains. (3) There is no proportionality between the acoustic emission and the mass of the material. (4) The thermal hysteresis can be associated with the delay in the reverse transformation caused by pinning of the martensite.

INTRODUCTION

The acoustic emission (AE) in the martensitic transformation of the Cu–Zn–Al alloys is progressively being studied in more detail, in order to relate it quantitatively to the kinetics and thermodynamics of the transformation [1–9]. The analyses have been performed using different methods to detect the AE: ring-down counting [3–7], amplitude distribution analysis [3,8,9], digital analysis of events [4] and multichannel acquisition which gives a high resolution in the distribution of counts by amplitudes [11]. In several experimental situations, the AE has been evaluated quantitatively [8,9]. A

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comparison between the duration of the acoustic activity and the time required to complete the transformation has suggested [3] the introduction of an "incubation period" for the activation of new sources. Generally speaking, the previous works globally study the transformation as a whole, in a temperature interval of about 30 K.

However, the problem of relating the thermodynamics of the transformation with the acoustic emission requires a previous knowledge of the dynamic response of the material–detector system. This can only be carried out following the theories of signal processing, from situations actually equivalent to the transformation itself. The relation is increasingly difficult as the signal-to-noise ratio becomes more important. The usual acquisition devices are designed for signal-to-noise ratios of between 10 and 100 [3,8,9]. Only recently has it been observed that the signal-to-noise ratio of the AE bursts may take a value above 1000 [12].

In addition, recent works exploring the possibilities of the heat-conduction calorimetry coupled simultaneously to other experimental techniques (acoustic emission, resistivity) have found some difficulties in the interpretation of the acoustic emission [7]. For example, disymmetries between the AE and the thermal power output have been observed. This is the reason why we are at present developing systems which are able to perform microscopical observations, either optical [13] or by scanning electron microscopy [14], simultaneously with measurements of the thermal energy, the acoustic emission, etc., of the transforming domains.

In this paper we present the results corresponding to a study, by coupled optical microscopy and acoustic emission detection, of the thermally induced martensitic transformation of a Cu–Zn–Al alloy under a mechanical applied stress. In this way, a single variant of martensite is produced, and, by varying the external load, the temperature at which the transformation begins can be adjusted to the neighbourhood of room temperature. This arrangement largely simplifies the experimental technology.

Our results establish the reproducibility of the AE related to the direct/reverse transformation cycles of the first martensite domain and the effects introduced by deviations in temperature around the domain of work. From the results obtained, the nature of the processes producing the acoustic emission can be ascertained.

EXPERIMENTAL

An optical microscope Olympus BH-2 with video-screen (maximum amplification $\times 700$) was used for the optical observations. The sample was attached to a copper-plate containing a Pt-100 thermoresistance. The copper-plate was then placed on a flat Peltier thermobattery used to cool/heat the whole stage of the microscope (see Fig. 1). The sample was

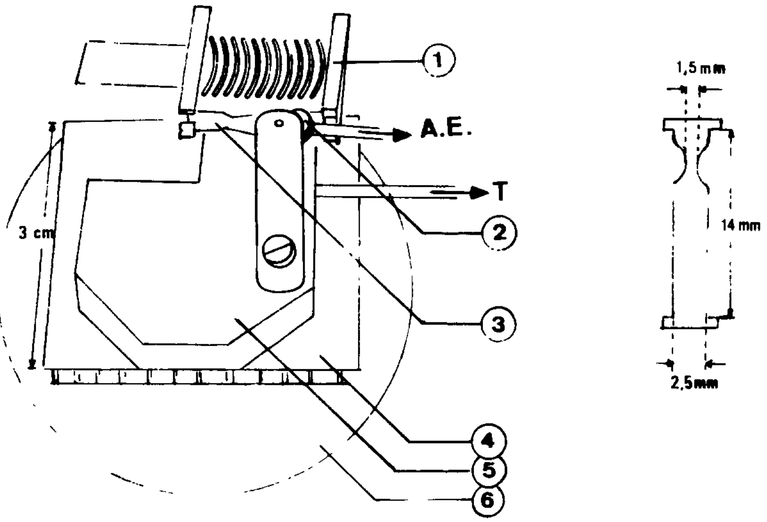

Fig. 1. Left: cooling/heating stage: (1) stressing device; (2) acoustic transducer; (3) sample; (4) Peltier thermobattery; (5) copper substrate; (6) copper block. Right: Sample shape and dimensions.

under tension by means of a spring. The acoustic signal was sent to a computerized digital acquisition system, as described previously [11] (piezoelectric transducer Philips, preamplifier and amplifier Bruel & Kjaer and multichannel Canberra 35, microprocessor EINA of 64K).

The sample used was a single crystal of 68.5Cu–14.9Zn–16.16Al (at%) 14.9; Zn, 16.6; Al 00.0 at%) with a uniaxial applied stress. The tensile axis was found to be at 10° from the [100] direction towards the centre of the fundamental triangle in the stereographic projection. Figure 1 shows its shape and dimensions. It was annealed in the β phase at 800°C for 20 minutes, and cooled in air to room temperature and, finally, it was electrolytically polished.

The acoustic emission measurements were performed using a total amplification of 74 dB, in the limit imposed by the signal-to-noise ratio for the sample and the experimental set-up used. The noise level after amplification centres around 50 mV. Usually, when studying global transformations an amplification of only 40 dB results in signals of more than 10 V. Qualitative studies of acoustic emission of this alloy (including the complete process of direct and reverse transformation) give signal-to-noise ratios above 80 dB. The surface area under study, where the microplates are produced, was restricted to ensure that the acoustic sources were very near to each other. In this way, even if there are important echoes of acoustic emission, the results can be related significantly to the processes taking place during the direct/reverse transformation of the martensite microplates. In addition, the material is free from adhesives, bonds and other constraints in the transforming area.

Using the multichannel apparatus in the MCS mode (multichannel swapping) the rate of events as a function of time at different scanning rates can be measured. In several instances, the acoustic activity was classified by amplitude, using the PHA mode (pulse height analysis) to evaluate its relative energetic importance.

The control of temperature in cycling was performed manually, with an uncertainty of $\pm 0.05\,^{\circ}C$ when reversing the processes of heating/cooling. The temperature was measured using a platinum resistance (Pt-100), with a resolution of 0.01 K. Cycling was done with mean heating/cooling rates of below 1 K min^{-1}.

RESULTS

Acoustic emission in the β-phase stability region (AEBP)

The noise level in the MCS mode was adjusted (without Cu–Zn–Al sample) to be near to one-half count/channel (in 2 s). A characteristic spectrum of this noise is shown in Fig. 2(A).

After introducing the specimen between the transducer and the copper substrate, the following was observed. (a) AE signals were detected while heating the sample in the β-phase stability region. The activity of this AE was more or less independent of the temperature, and it was observed to be at least up to 100°C above the Ms temperature (birth temperature of microplate on cooling). This AE activity was considerably lower on cooling than on heating in the same temperature domain. Similar behaviour was observed either for free or stressed material. (b) When the sample was thermally cycled within a temperature interval of about 4 K, the AEBP decreases with the number of cycles [see Fig. 2(B)]. In some cases, cycling shows a reproducible residual pattern. If the width of the temperature interval was reduced to less than 2°C then the AE almost disappears after the first cycle [see Fig. 2(C)].

The AE in the transformation / retransformation of a martensite microplate

The stressed specimen was cooled down from the β-phase stability region until small plates of martensite appeared in the neck shaped area. Most of the plates were found to start from the edges (lateral surfaces). Trace analysis indicated that the shear directions of the variants formed were closely parallel to the lateral surface and inclined by about 45° with respect to the normal of the horizontal surface. In order to isolate only one martensite plate, the sample was cooled, heated or its position in the

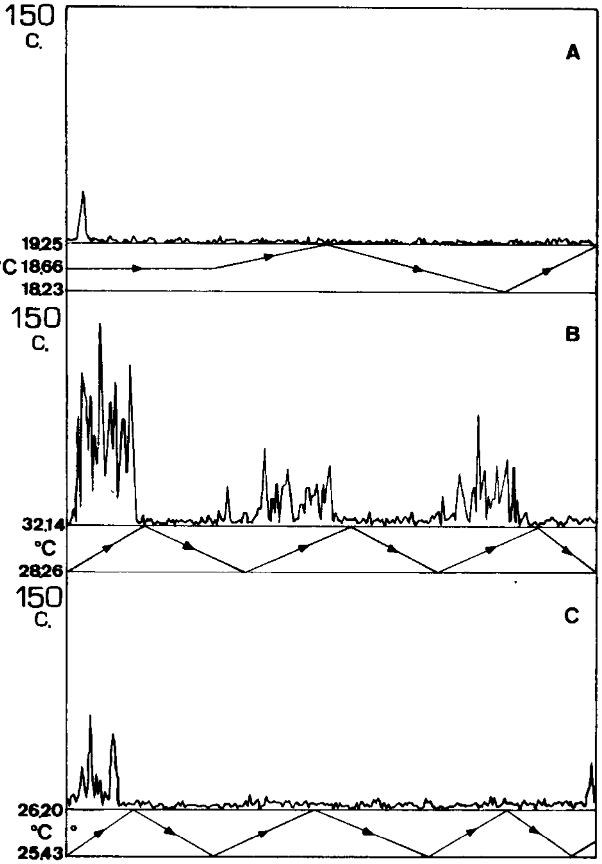

Fig. 2. Acoustic emission spectra corresponding to: (A) noise level in absence of sample; (B) AE in the β-phase stability region decreasing with cycling in a temperature interval of $\Delta T \simeq 4$ K; (C) AE in the β-phase stability region almost disappearing after the first cycle with $\Delta T \simeq 2$ K.

stressing device was modified as many times as necessary. Meanwhile, it was observed that any AE that could be associated with the transformation/retransformation of a martensite plate was comparable to the AEBP in such a way that no clear connection between this AE and the movement of the plate could be established. Under this circumstance it was found necessary to reduce the width of the temperature interval to less than 2 K.

Nevertheless, in this small temperature interval, it was found to be possible to obtain a microplate of martensite with a reproducible appearance/disappearance behaviour on cycling. The dimensions of the microplates obtained in this way were typically: length < 400 μm, width < 3 μm, and height < 500 μm. An example is shown in Fig. 3. Although some thickening occurs, the growth of the martensite plate is essentially longitudinal; i.e., in a direction parallel to the habit plane.

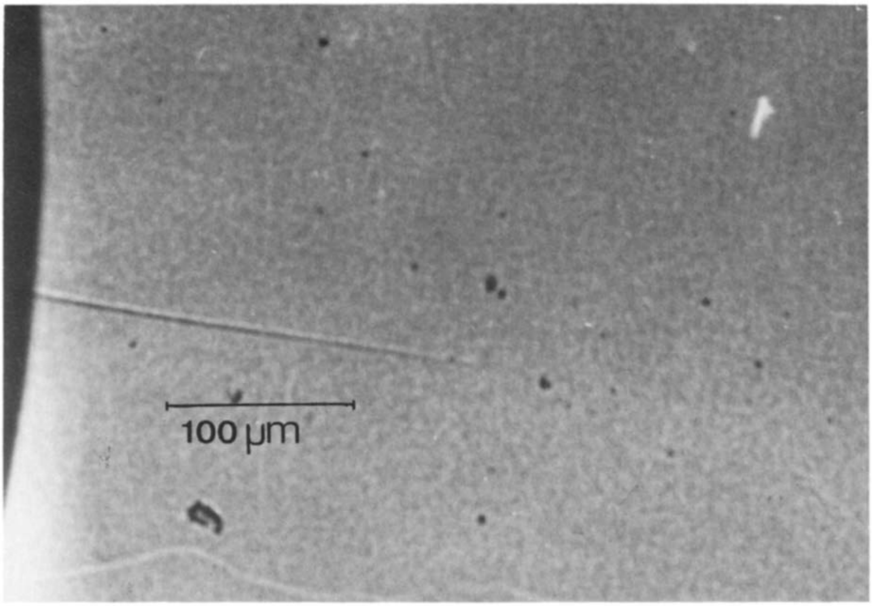

Fig. 3. Micrography showing an isolated martensite plate.

Under this condition, several isolated martensite microplates were cycled, and the AE was recorded either in the MCS or in the PHA mode of the multichannel. The results are described below.

(a) The longitudinal transformation of a martensite microplate proceeds more smoothly and in a shorter time (less than 1 s) as compared with the retransformation. Occasionally, some segments of the microplates were observed to grow suddenly. An AE signal was detected associated with this more accelerated growth.

(b) In most cases the martensite microplates were found to shrink by steps on heating. When the microplates moved slowly, no AE was detected, but when a segment of the martensite suddenly shrank, then an AE peak was observed. In particular, it should be emphasized that the disappearance of the last segment of the microplate was, in most cases, associated with the highest AE activity. The duration of the whole disappearance process was; typically, of the order of 20–30 s. A qualitative hysteresis curve of the martensite length (l) as a function of temperature (T) is shown schematically in Fig. 4.

(c) Successive cycles in the same temperature interval indicated that an AE spectrum with a reasonable reproducibility could be obtained. Two examples are shown in Figs 5 and 6. Figure 5 corresponds to the case in which an AE peak was detected during the transformation, and is clearly associated with an acceleration in the growth of the microplate. Several peaks were observed on heating, but the only ones clearly related to the

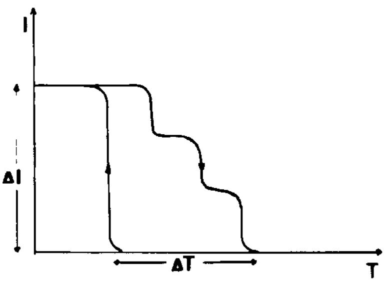

Fig. 4. Qualitative hysteresis curve of the martensite plate length (l) as a function of temperature (T). $\Delta l \simeq 250$ μm, $\Delta T \simeq 0.5$ K.

movement of the plate are those labelled d1 and d2. Figure 6 shows an AE spectrum in which no peaks were observed during the transformation, but several peaks were recorded as the plate shrank on heating. Among these peaks, those labelled d1 and d2 were clearly associated with a sudden movement of the plate. The other peaks, indicated by arrows, were not related with any visible movement of the plate; they are probably due to the AEBP.

(d) The results corresponding to several microplates are summarized in Table 1. The AE activity has been represented by the means of and standard deviations in the number of counts detected by the multichannel. In this study, the stability of temperature programming (hand controlled) was not great (±0.05 K). The partial instability of the microplate was, probably, therefore, related to the fluctuations in temperature. Small changes in temperature or, obviously, stressing, induce new microplates and/or changes in the acoustic emission pattern.

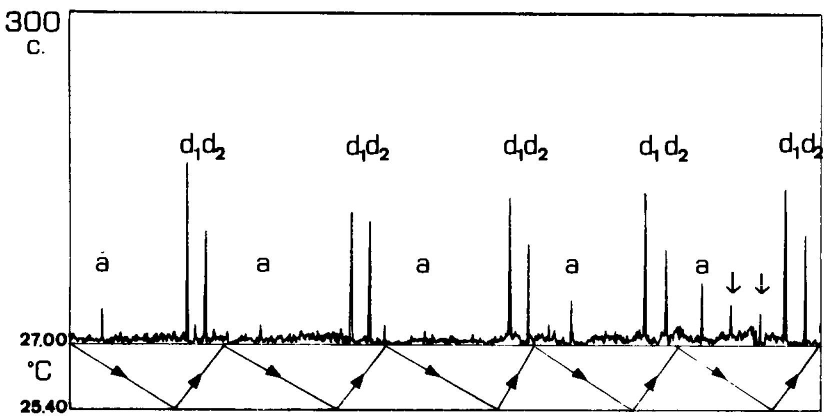

Fig. 5. Acoustic activity vs. temperature in successive cycling of an isolated martensite plate. (a) The appearance of the plate; (d1 and d2) the shrinkage of the plate. Other peaks that cannot be associated with any visible change in the plate are indicated by arrows.

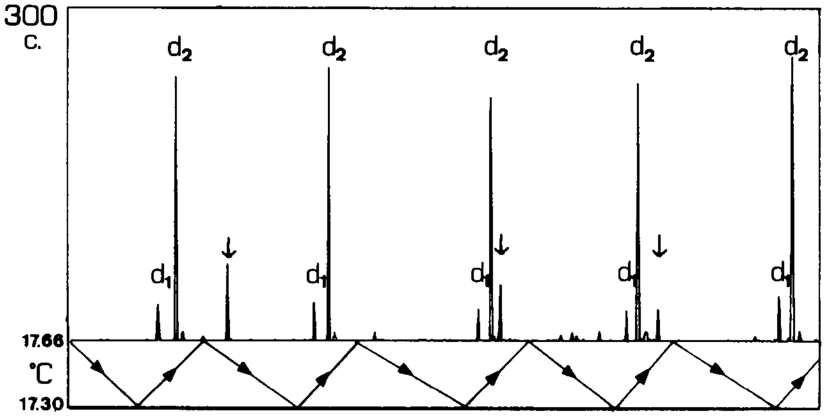

Fig. 6. AE activity vs. temperature in successive cycling of an isolated martensite plate in the case in which no AE signals are detected during the appearance of the plate. The peaks labelled d1 and d2 clearly correspond to sudden movements of the plate, while those indicated by arrows are not related to any visible movement of the plate.

TABLE 1

Acoustic emission activity for several microplates [a]

Appearance			Disappearance			Hysteresis	
N	Ms	NA	Af	NI	NF	$\Delta T = \mathrm{Ms} - \mathrm{Af}$	n
1	26.02±0.05	34±10	26.71±0.07	186±18	99±10	0.69±0.04	5
2	17.35±0.01	0	17.60±0.01	35± 4	239±13	0.25±0.02	5
3	19.94±0.01	0	20.55±0.04	106±40	305±23	0.61±0.04	8
4	19.79±0.01	0	17.99±0.01	42±10	288±13	0.20±0.01	6
5	18.71±0.04	86±18	19.41±0.02	177±33	727±33	0.70±0.01	6

[a] N, microplate number. Ms, birth temperature of the microplate on cooling (°C). NA, number of counts in the appearance peak. Af, temperature of disappearance of the last trace of martensite on heating (°C). NI, counts of all the intermediate peaks on heating; NE, counts of the last peak in the retransformation. n, Number of cycles studied.

(e) The time width of the peaks are typically about 0.6 ms, except for the last peak in the disappearance which can be up to 2.5 ms wide. An analysis of this peak shows, in some cases, an internal structure formed by two or more discontinuous signals.

(f) The acoustic emission study, via the PHA mode, shows an amplitude ratio between transformation/retransformation of near 1/4. This result is in agreement with global observations of acoustic emission.

DISCUSSION

The pinning of the martensite microplates

The fact that, in most of the observed cases, the motion of a microplate occurs by steps, suggests that pinning centres exist in the material. These pinning centres behave asymmetrically for the transformation and the retransformation. They have very little affect on the transformation (smooth longitudinal growth) but they retain the microplates during the retransformation until the necessary thermodynamic energy required to surmount this barrier is accumulated. This excess of thermodynamic energy produces a more accelerated shrinkage of the microplate, thus giving rise to AE.

Baram and co-workers [3] studied the acoustic emission from a single-interface martensitic transformation in an Au–Cd alloy. They found that no acoustic emission was detected during the transformation, except when twinned bands appeared in the sample. Conversely, AE was invariably recorded during the retransformation. Although the experimental procedure of Baram and co-workers was different from ours, it is interesting to note that the global results are qualitatively analogous.

It was not possible to obtain any information about the nature of the pinning centres from our experimental results. Nevertheless, it seems reasonable to associate them with relatively isolated defects or defect packages present in the material. Such defects can be pre-existent in the β phase (dislocations, aniphase boundaries, vacancy clusters, surface defects, etc.) or be generated by the transformation itself. More research is needed to elucidate this point.

Surface effect over the martensite microplate movement

It was suggested by Delaey et al. [15] that the tip of the 18R martensite (at least in thin foils) can be thought of as a screw dislocation with a Burgers vector (**b**) parallel to the macroscopic shear direction, whose magnitude is given by:

$$\boldsymbol{b} = se \tag{1}$$

where s is the amount of the macroscopic shear (~ 0.1) and e is the thickness of the plate.

In the particular case studied in this work the shear direction of the transforming martensite plates was, approximately, parallel to the lateral faces of the sample. If the martensite microplate is thin enough, then the total deformation that it produces in the surrounding β-phase is approximately equivalent to that corresponding to a screw dislocation with the Burgers vector given by eqn. (1), and located at the tip of the microplate. Under this circumstance it could be expected that the martensite will behave like a screw dislocation in the vicinity of the surface. This is illustrated in Fig. 7.

Assuming an isotropic medium, the screw is drawn toward the surface by a force [16]:

$$Fx/L = \mu b^2/(4\pi x) \tag{2}$$

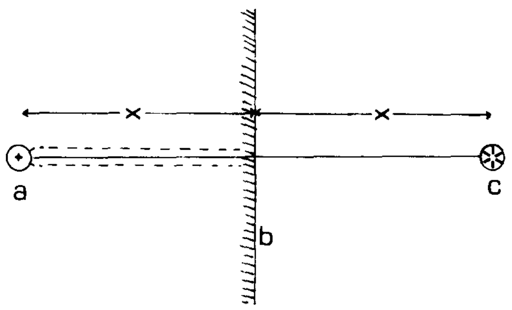

Fig. 7. Schematic diagram of the surface effect on the martensite microplate: (a) screw dislocation (martensite microplate); (b) surface; (c) image screw.

where L is the length of the dislocations and μ is the shear modulus. Therefore, in this scheme, as the microplate shrinks toward the surface it will be increasingly accelerated [as it can be deduced from eqn. (2)] until it annihilates against its virtual image. This surface effect could explain why, in most cases, the highest AE activity is detected at the last disappearance of the microplates. Such a surface effect is also a possible explanation for the lack of AE during the formation of a martensite microplate, because the surface attraction can prevent a high accelerated growth of the plate.

The nature of the AE from a martensite microplate

It is well known that the martensitic transformation produces a shape change of the sample. The appearance of a small plate of martensite produces elongation (or shortening) of part of the sample; this is illustrated in Fig. 8. The edge AD is longer (or shorter) than the edge EF. Moreover, the sample between AB is rotated vs. CD. In addition to this rigid movement, a sudden appearance/disappearance of a martensite plate will produce elastic vibration along the sample. Both phenomena can act on the transducer giving rise to AE signals.

In these conditions quantitative results of acoustic emission are related to several parameters, which can be intrinsic or extrinsic to the characteristics of the transformation; these are given below.

(a) Intrinsic parameters: the AE depends on the length, number of steps and acceleration in each step during the movement of a plate. These

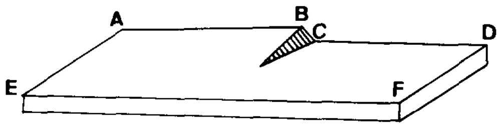

Fig 8. Shape change produced in the sample by the appearance of a martensite plate.

processes in turn are determined by the interaction of the martensite with defects, surfaces, etc. They will also depend on the thickness and crystallographic orientation of the plate with respect to the surface of the foil.

(b) Extrinsic parameters: the acoustic emission would depend on the shape and size of the whole sample (through the normal vibration modes). The applied stress can also have some influence.

In addition to these factors, the response of the detection system should be known in order to obtain a quantitative relation between the AE spectrum and the intrinsic parameters of the transformation. Nevertheless, if all these factors remain constant, the relative difference between the AE from two or more events which are close to each other can be a very useful information.

The acoustic emission from the β phase (AEBP)

The AEBP shows a similar behaviour to that of the martensite microplate transformation; i.e., the AE activity is higher on heating than on cooling and, moreover, some at least partially reproducible patterns can be obtained on cycling. This suggests that the AEBP is closely related to the martensitic phenomena. In this respect, the presence of surface martensite in this material should be taken into account. Surface martensite (SM) is found in the β phase of most noble metal-based alloys. The main characteristics of the SM are [17,18]: (a) it is present at least up to ca. 500 K even for alloys with very low Ms or which do not transform martensitically at all; (b) its structure and number of variants depend on the surface orientation, and no SM is observed in $\{100\}$ surfaces; (c) the thickness of the surface martensite is of the order of 10 nm for flat surfaces, but it can be larger at the edges or irregularities of the surface.

Fluctuations of the SM can be a plausible source of AE, even when the bulk material is in the β phase stability region. The fact that the AEBP disappears when the material is cycled in a temperature interval of $\Delta T \simeq 1$ K, suggests that the hysteresis for the movement of any SM domain is wider than this ΔT.

CONCLUSIONS

(1) The signal-to-noise ratio has been enhanced to explain, qualitatively and semiquantitatively, the global features of AE in martensitic transformations. The intensity of fine structure vs. high-intensity pulses ratio during the transformation is near 1/1000.

(2) Acoustic emission from the β phase (AEBP) was detected up to temperatures far above that of the martensitic transformation. Study of thermal cycling shows two remarkable features: (a) suppression of AEBP

with a thermal expansion of $\Delta T < 2$ K; (b) drastic reduction of AEBP for $\Delta T \simeq 3$ K and, in some cases, reproducible pattern of isolated acoustic pulses.

(3) Acoustic emission in thermal cycling ($\Delta T < 2$ K)– longitudinal transformation/retransformation– of the first martensite microplate shows that: (a) when the microplate moves smoothly no acoustic emission is detected, and only in sudden motions (accelerations) is AE activity measured; (b) the AE activity is lower in the $\beta \to$ martensite than in the martensite $\to \beta$ processes; (c) the acoustic emission proceeds in burst-steps (AE plus echoes < 600 μs); (d) the last burst (last microplate-step in the retransformation) has, normally, the greatest counting, and also the time-expand of the activity is increased (> 1 ms).

(4) Reproducible results were obtained in the following observations and measurements: (a) optical observations; (b) AE activity (global and burst pulses); (c) transformation/retransformation temperatures, hysteresis and burst processes.

(5) Some remarkable features of the martensitic transformation are proposed: (a) the AEBP is tentatively attributed to the presence of surface martensite; (b) the AE is associated with acceleration in the martensite microplate growth/shrinkage, i.e., local phenomena related to pinning centres; (c) the AE is not mass connected. The relation between the AE and the transformed mass is statistical via the pinning centres; (d) the hysteresis in the longitudinal transformation/retransformation process is mainly determined by the pinning of the microplate during the retransformation.

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