CHARACTERIZATION OF PHASE EVOLUTION UNDER LOAD BY MEANS OF PHASE CONTRAST IMAGING USING SYNCHROTRON RADIATION

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ABSTRACT
In the last few years many studies have been done on the characterization of the phase evolution, during martensitic transformation of shape memory alloys (SMAs). From the experimental point of view this topic has many difficulties. Detailed information is needed on the very small modifications locally induced by the transforming interface and, at the same time, on the phase evolution in the sample as a whole. Microscopic techniques give detailed information on local interfaces, twinning and self-accommodation structures, but in order to develop a complete description of the link between the microscopic and macroscopic levels, proper techniques must be identified and verified which can describe the intermediate level.

In this paper we report on the application of Phase Contrast Imaging (PCI) in the study of the phase evolution during pseudoelastic transformation in NiTiCu shape memory alloys. PCI is a quite novel technique, which gives image information linked to very small differences in the density of the material under analysis. The method has some clear advantages when compared with common microscopic techniques: (a) no special preparation of the sample is needed, (b) the investigated area is very large, (c) it allows the setting up of complex experimental apparatus.

In order to perform stress-strain tests on NiTiCu samples, an "ad-hoc" experimental set-up was prepared. With this, control of different parameters and data acquisition during tests were ensured by dedicated software, developed in the LabVIEW language, while images of the sample were acquired.

INTRODUCTION
The interest in SMAs has been increasing owing to the functional properties of these materials such as pseudoelasticity and Shape Memory Effect. These effects are of interest both for basic research and for the development of new applications. Many investigations were undertaken on the most common shape memory systems (e.g. NiTi, CuZnAl) with the aim of understanding the complex relation between the microscopic and macroscopic behaviour of these materials. In a successful experiment, a thermal imaging technique has been used for detecting small local temperature variations due to latent heat of transformation [1].

Another successful approach was the characterization of Cu-Zn-Al single crystals

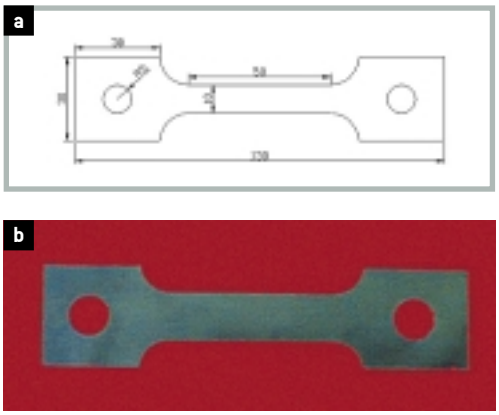


Figure 1. a) schematic view of the sample laser cut from the rolled foil. The sample thickness was 0.1 mm. **b)** is a photograph of the real sample. The two holes have been cut to increase connection in the grips.

[1] A_F =Temperature of the end of the austenite transformation

M_F = Temperature of the end of the martensite transformation

by using synchrotron X-ray diffraction topography technique [2].

In this paper we present a characterization of phase evolution under load by means of synchrotron radiation PCI. The high transverse coherence of radiation delivered from third generation synchrotron light sources like ELETTRA allows us to overcome the usual restriction of radiography to absorption-induced contrast, and to visualize object features that affect the phase of a transmitted X-ray beam. In this imaging mode, the sample is illuminated by a monochromatic hard X-ray beam, and a position-sensitive detector is set at a distance typically between 1 cm and a few m from the specimen. Free space propagation transforms the phase modulation of the transmitted beam into an amplitude modulation [3]. Contrast basically stems from interference between parts of the wavefront that have experienced different phase shifts. The technique has been demonstrated to be useful both for the characterization of very thin biological structures as well as of light metals [4] and metal matrix composites [5].

In this case we used PCI in an attempt to identify the growth mechanism of Stress Induced Martensite (SIM) during mechanical characterization of a $Ni_{45}Ti_{50}Cu_5$at% alloy in the pseudoelastic regime.

EXPERIMENTAL

We used $Ni_{45}Ti_{50}Cu_5$ at% prepared at CNR-IENI laboratories. We prepared, by hot rolling and cold rolling with intermediate anneals, a thin ribbon (25x0.1 mm^2) with residual cold deformation (estimated as thickness reduction) of about 30%. Small pieces (about 200 mm in length) were submitted to a thermal treatment of 1.8 Ks at a temperature of 723 K. Owing to geometrical constraints imposed by the special apparatus described below, a sample like that shown in Figure 1 was laser cut from the thermally treated piece.

The samples were electropolished in order to eliminate surface oxide, formed during the thermal treatment.

The transformation temperatures of the sample were assessed by means of calorimetric characterization. For this purpose a Seiko DSC 220C calorimeter, equipped with a liquid nitrogen stage, was used. The analysis were performed at a scanning rate of 5 °C/min over the temperature range -50 °C / + 150 °C. The calibration of the temperature scale was obtained by comparison with the melting point of pure elements: In and Hg.

In order to perform stress-strain tests an "ad-hoc" experimental set-up was prepared. A schematic view of the apparatus is reported in Figure 2. During tests several parameters can be measured: strain by means of a Linear Variable Differential Transformer (LVDT)[1], force by means of a loading cell, electric resistance by means of four probes directly mounted on the measuring sample. The entire experimental set-up was mounted in the hutch of the SYRMEP beamline and a sequence of images during a complete pseudoelastic cycle was acquired by means of photographic plates or a 2048x2024 pixels CCD detector (pixel size = 14 μm).

First of all PCI was applied to a stress-free sample which underwent only a thermal transformation. Images of the sample at a temperature well above A_F were acquired. They were compared with images of the same specimen area at a temperature well below M_F. In this way it was possible to appreciate the differences between the two images that are related to the martensitic transformation. This procedure allowed us to identify the best experimental parameters for PCI.

Afterwards a complete pseudoelastic cycle was performed. Both during the loading and unloading of the branches the cycle

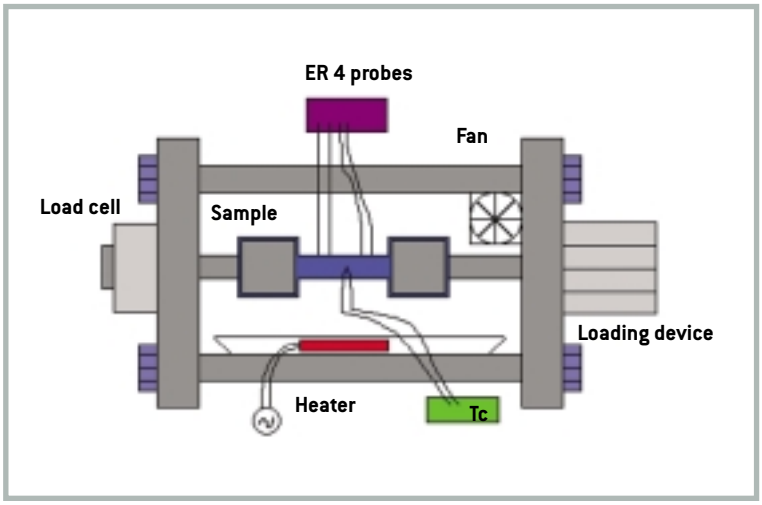


Figure 2. Schematic view of the special apparatus developed to perform PCI characterization. The whole apparatus has a maximum length of 230 mm and a maximum width of 150 mm.

FIGURE 3

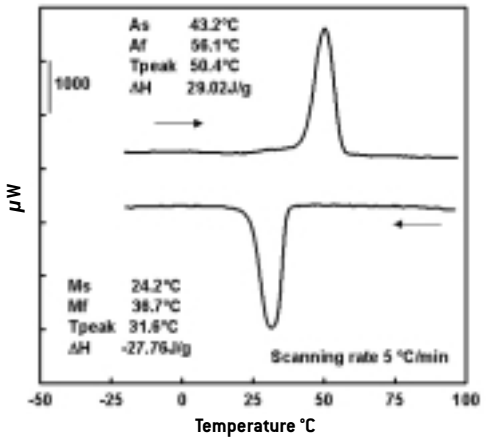
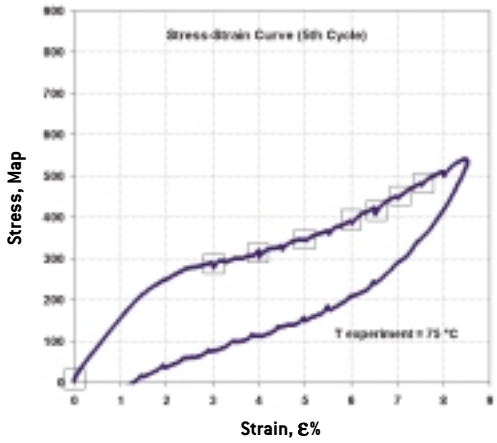


FIGURE 4



was stopped at specific points (e.g. pseudoyield point, half value of the transformation strain, end of the pseudoelastic plateau) and a radiograph was taken. The pseudoelastic cycle was measured at a temperature of 20 °C above the A_F temperature.

RESULTS AND DISCUSSION

Figure 3 shows a typical thermogram of the material. Figure 4 is the stress-strain measurement performed on the sample mounted in the special device at a test temperature of 75 °C. While the calorimetric curve is the one expected, the stress-strain one deserves some comments. It was not possible to observe a pseudoelastic cycle with a straight loading plateau. In any case, it is possible to recognize two slope changes which can be related to the start of the stress induced transformation (at about 2% of strain) and to the onset of elastic deformation of the SIM (roughly at 6-7% of strain).

In order to evaluate the resolution limit of PCI and the influence of the surface of the specimen, some preliminary tests were performed. First of all a comparison was conducted between the images taken by conventional optical microscopy and the phase contrast technique. Preparing the sample according to the needs of optical microscopy [6] is detrimental for PCI. In fact chemical etching has the effect of increasing the evidence of surface structures. As a consequence it is much more difficult to appreciate bulk modifications due to the transformation. An example is reported in Figures 5 a) and b).

The fine structure which is evident in the optical micrograph, is present even in the PCI image but it is so fine and dispersed that other structures are blurred. Without chemical etching, bulk features related to the phase transformation are easily identified. One example is reported in Figure 6.

Here the same sample area (the white spot visible at bottom right of the two photos is a reference mark) has been photographed in the two phases, fully austenite ($T>A_F$) and fully martensite ($T<M_F$). The effect of the transformation is observed by formation of a fine dispersed structure which is shown in Figure 6 b). It is worthwhile to recall that the observed structure is not related only (as in the case of optical microscopy) to the formation of surface relief, but is directly related to the bulk structure of the transformed sample. When the specimen was observed at different temperatures without any chemical etching no visible modification of the surface was detected.

The structure shown in Figure 5 b) is not changed by changing temperature. Assuming that this is the real structure of martensite and not an artefact due to chemical etching one must conclude that this is stabilized at the surface so that no information on temperature related phase evolution can be drawn from it.

According to the parameters obtained by this preliminary investigation a complete

Figure 3. Calorimetric characterization of the sample used for PCI characterization. The main transformation parameters are reported in the graph.

Figure 4. Typical stress strain curve as measured in the sample installed in the special apparatus. Boxes highlight the strain values at which the phase contrast image was acquired.

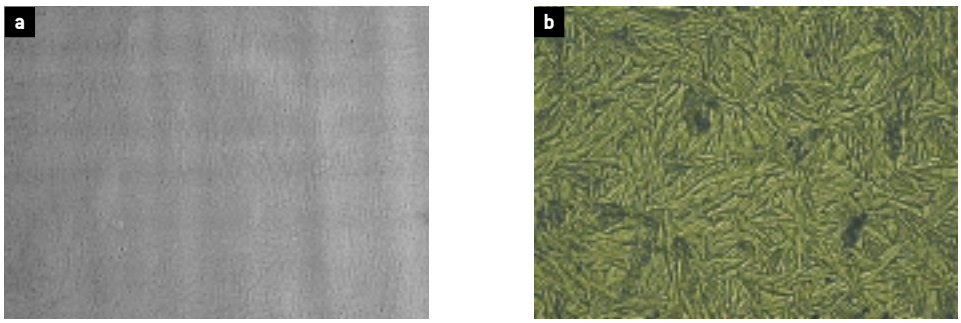


Figure 5. a) PC image of a sample with the surface prepared for optical microscopy observed in phase contrast; **b)** optical microscope image of the surface of the sample. See text for details.

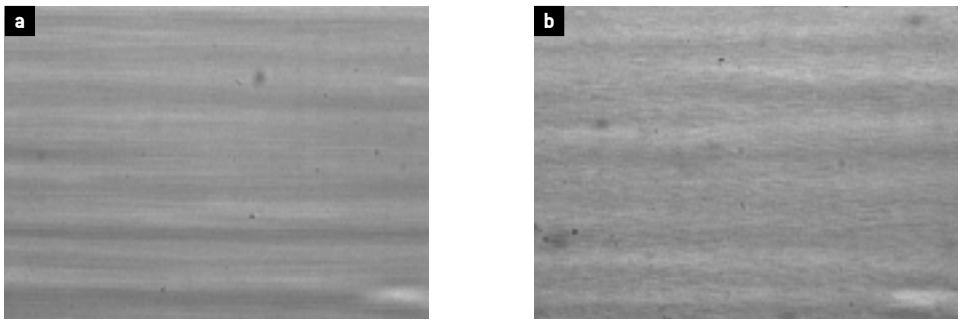


Figure 6. PC images of the NiTiCu sample in the austenitic phase at $T>A_F$ **a)** and in the martensitic phase **b)**. The two images have been taken from the same sample area using as reference the white spot, bottom right.

pseudoelastic cycle has been characterized by PCI. On the basis of the mechanical characterization performed, some points have been identified (represented by boxes in Figure 4) at which the PCI observations were performed. In the sequence of photographs of Figure 7 the images taken at different strain levels are compared. To better appreciate the modifications induced by increasing strain, photographs have been post-processed.

In each image, some lamellae are present. For low values of the strain (say up to 4-5%) they do not change in shape or in apparent intensity. At a deformation of about 6% there is an increase in the intensity (the lamellae are darker, i.e. they are more radiation-opaque). From about 6.5% they change slightly in shape, at 7% they are clearly thinned and, eventually, at 7.5% there is no evidence of these lamellae.

The intensity variation due to lamellae evolution is underlined by the curve in Figure 8 where the intensity evolution versus imposed deformation is shown.

The lamellae which disappeared at the end of the loading, retransform during unloading. In fact they are still present at the beginning of the following cycle. Images reported in Figure 7 refer to the 5th complete pseudoelastic cycle to which the sample was submitted. As a consequence the behaviour can be considered quite reproducible.

CONCLUSIONS

The PCI technique proved successful even in the characterization of purely thermally activated transformations.

Interesting results were obtained in the characterization of a pseudoelastic cycle performing PCI during the formation of SIM. We have found that some structure exists which is stable up to a strain value which is similar to the measured maximum transformation strain. Once this value is passed, the structure disappears but is still present after unloading. Further work is needed to clearly correlate this behaviour to the properties of the underlying martensitic transformation.

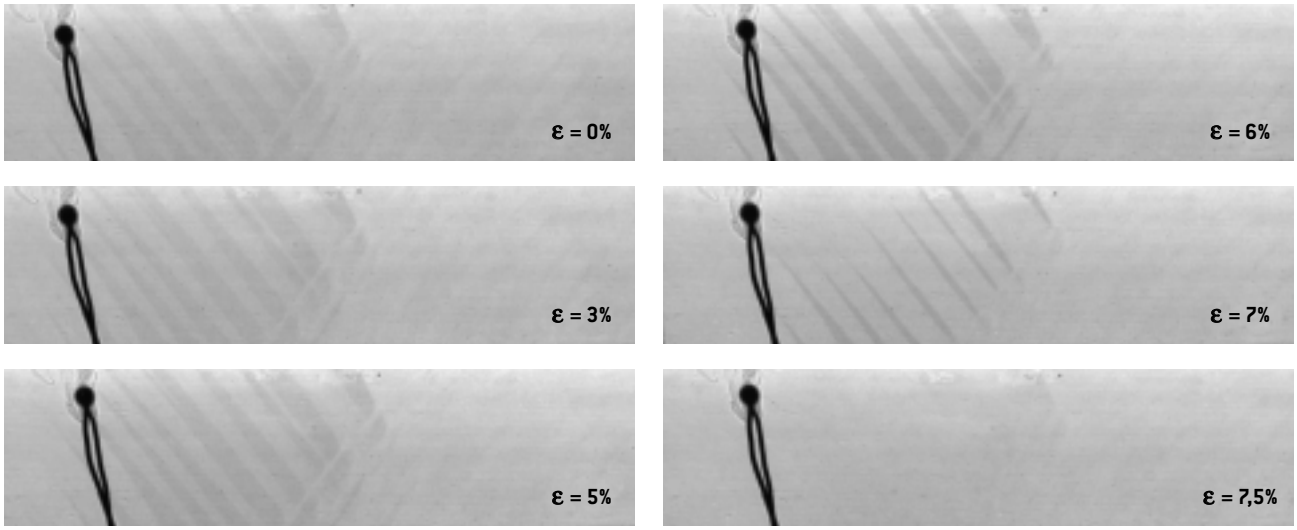


Figure 7. PC images of the sample at different values of imposed strain. The images have been processed in order to enhance the contrast contribution coming from the different phases.

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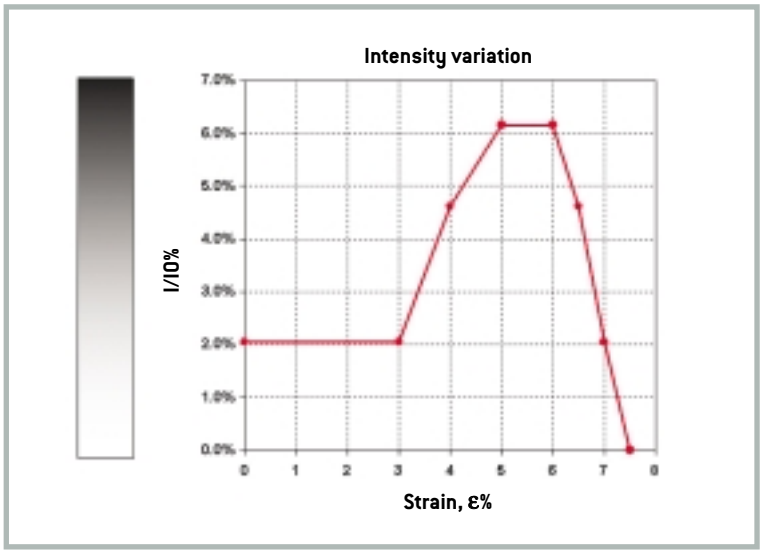

Figure 8. Intensity evolution of Lamellae vs. the imposed strain.