



PERGAMON

Scripta Materialia 46 (2002) 471–475



www.actamat-journals.com

# Stress-induced FCC/FCT phase transformation in Fe–Pd alloy

Hiroyuki Kato<sup>\*</sup>, Yuanchang Liang, Minoru Taya

*Department of Mechanical Engineering, Center for Intelligent Materials and Systems, University of Washington, Seattle, WA 98195-2600, USA*

Received 2 April 2001; accepted 22 August 2001

## Abstract

We report stress-induced martensitic transformation in tensile loaded Fe–30.5 at.% Pd polycrystals, mainly using optical microscopy. Reverse transformation occurs during unloading. Thermodynamic relationships between applied stress, transformation strain and the latent heat of transformation are examined. © 2002 Acta Materialia Inc. Published by Elsevier Science Ltd. All rights reserved.

*Keywords:* Ferromagnetic shape memory alloy; Phase transformation; Stress–strain relationship; Stress-induced martensite; Iron–palladium

## 1. Introduction

A topic of recent interest in martensitic transformation is concerned with ferromagnetic shape memory alloys [1–8]. In these alloys, the transformation and associated straining can, in principle, be controlled by a magnetic field. Mechanical actuation by a magnetic field is a useful application of this phenomenon. A magnetic field can supply work to the alloys for this purpose. Superelasticity and shape memory must be assessed to explore the usefulness of ferromagnetic shape memory alloys for magneto-mechanical applications.

Beginning with the work of Sohmura, Oshima and Fujita, an Osaka University group studied extensively thermoelastic martensitic transformation in Fe–Pd [9,10]. Both austenite and martensite in Fe–Pd are ferromagnetic. Even though thermoelasticity implies the existence of stress-induced transformation and the shape memory effect, these properties have not yet been fully examined in Fe–Pd. Fe–Pd exhibits lattice softening: Young's modulus decreases as temperature is lowered towards  $M_s$  [11,12]. If stress-induced transformation occurs, the additional strain from the transformation reduces overall stiffness. Low stiffness is advantageous for an application that requires a large displacement from a small load. This paper presents evidence for stress-induced martensitic transformation (SIM) in Fe–Pd. The transformation is discussed on the basis of thermodynamics and geometrical analysis.

<sup>\*</sup> Corresponding author. Permanent address: Mechanical Science and Engineering, Hokkaido University, Sapporo 060-8628, Japan. Tel.: +1-206-685-2850; fax: +1-206-685-8047.

*E-mail address:* tayam@u.washington.edu (H. Kato).

## 2. Experimentals

The present study examines the FCC to FCT martensitic transformation of Fe–30.5 at.% Pd polycrystals. This alloy has an onset transformation temperature,  $M_s$ , just below room temperature [10–13]. An Fe–Pd ingot having this composition was made of granular iron (99.98%) and palladium powder (99.95%), which were melted in an arc furnace. The ingot was first homogenized and then cold rolled into a 1 mm thick sheet. Shaped specimens were annealed at 1250 °C for 12 h in vacuum and quenched into iced water. Some specimens were electrolytically polished for metallographic observations. The martensitic transformation was confirmed by optical microscopy, electrical resistivity measurements and calorimetry (differential scanning calorimetry, DSC, Perkins–Elmer). Tensile tests were performed on an Instron type machine.

## 3. Results and discussion

Transformation temperatures,  $M_s$ ,  $M_f$ ,  $A_s$  and  $A_f$ , determined by the resistivity measurements were 2, –9, –7 and 5 °C, respectively. The transformation temperature determined by optical microscopy varied from grain to grain, presumably due in part to the inhomogeneity of composition: in particular,  $M_s$  depends strongly on the composition,  $\sim 10^2$  °C/at.% Pd [10]. In spite of macroscopic fluctuation from grain to grain, the hysteresis observed in individual grains was very small compared with the reported value of 10 °C in single crystals [13]. For example, two martensite crystals in a grain nucleated at –0.6 °C grew across the grain at –0.9 °C, as shown by the arrows A and B in Fig. 1(a). At –2.9 °C the whole grain was covered by other martensite plates, (b). Upon heating, the two plates disappeared at 0.3 °C. Thus, the hysteresis of this grain is

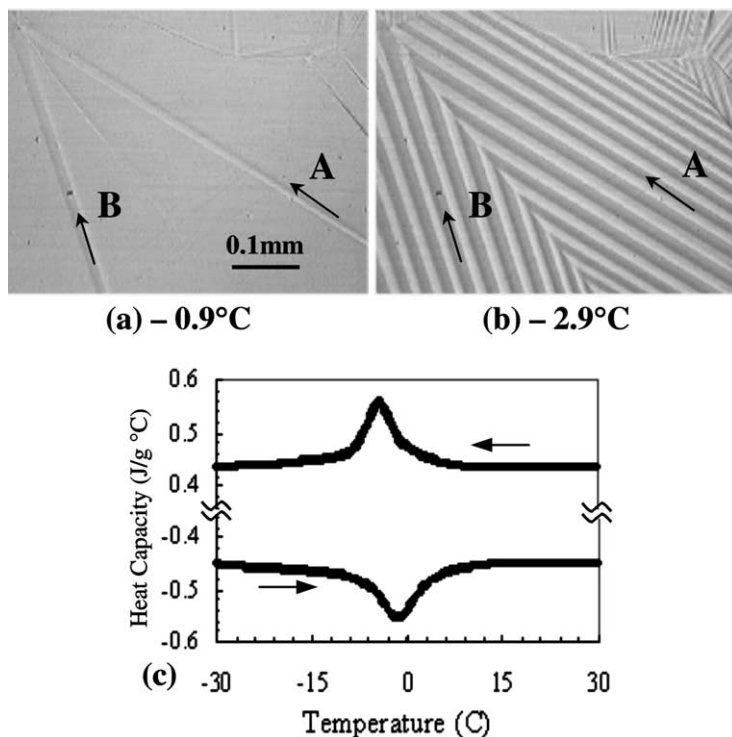


Fig. 1. (a,b) Optical structures. (c) A DSC curve.

0.9 °C. Other grains exhibited similarly small hysteresis.

Some researchers have reported that the martensitic transformation in Fe–Pd is nearly of second order [13]. However, the present study found the clear existence of latent heat of the transformation. Endo- and exothermic peaks in heating and cooling runs (DSC) shown in Fig. 1(c) give the latent heat of the transformation of 0.53 J/g. Thus, the transformation is of first order.

Stress-induced martensite was observed microscopically in several grains during uniaxial loading of a sheet specimen ( $60 \times 3 \times 1 \text{ mm}^3$ ) at several temperatures. The martensite structures, formed in a grain, are shown in Fig. 2. Fig. 2(a) shows for

martensite plates formed by cooling to  $-10 \text{ °C}$ . These plates disappeared during heating at a temperature of  $4 \text{ °C}$ . Martensite plates with bright contrast were formed when the specimen was stressed to 12 MPa at  $6 \text{ °C}$ , (b). As the stress was increased to 48 MPa, the width of the plates increased, (c). These martensite plates completely disappeared after unloading at  $6 \text{ °C}$ , (d). That is, as far as individual martensite plates are concerned, the stress-induced transformation is completely reversible. However, the strain caused by loading did not recover during unloading. Fig. 3(a) is the stress–strain curve of a specimen deformed at  $16 \text{ °C}$ . At points A, B and C, the specimen was unloaded and reloaded as indicated in the figure.

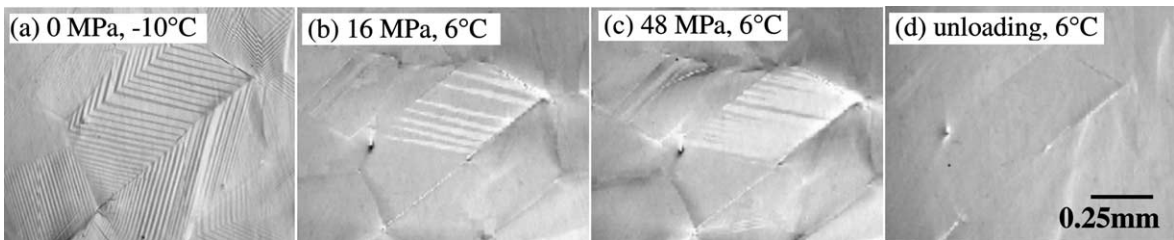


Fig. 2. Martensite formed by cooling (a) and formed by loading (b) and (c). Stress-induced martensite disappears upon unloading (d).

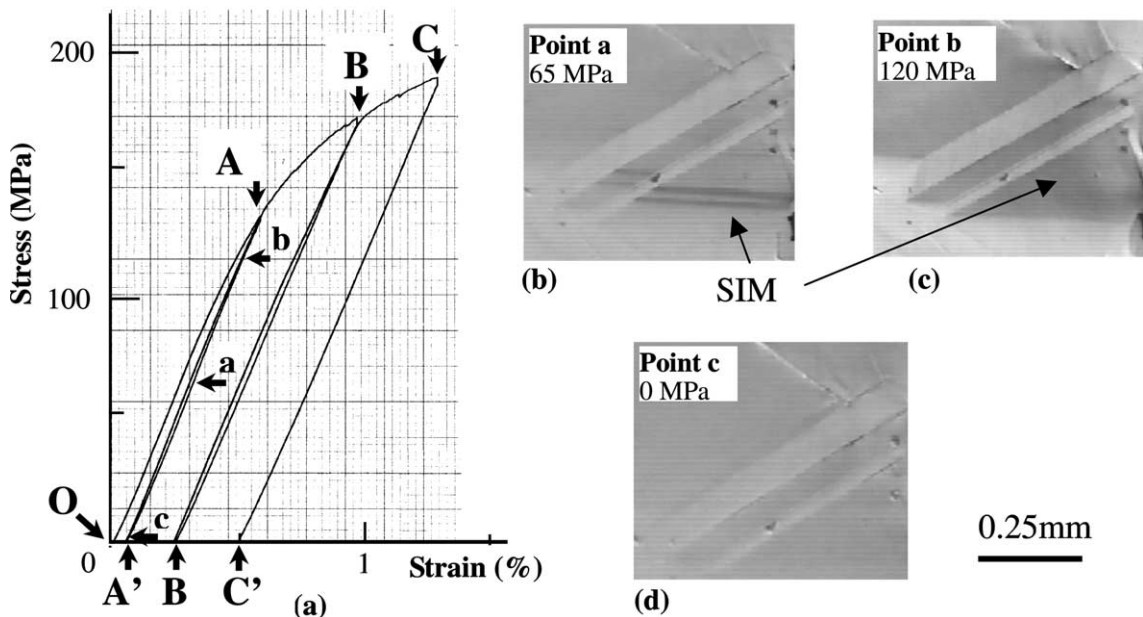


Fig. 3. (a) A stress–strain curve at  $16 \text{ °C}$ . (b)–(d) The micrographs taken along the path AA' shown in (a).

During these cycles, the martensite content increased with load, but the martensite plates, thus formed, disappeared completely upon unloading. An example is shown in Fig. 3(b)–(d) where (b) indicates the onset of twinned martensite plates (in dark stripe) which spread over the entire dark region, (c). Even though, the stress–strain curve in Fig. 3(a) does not show clearly superelasticity, the SIM occurred under increasing stress as evidenced by Fig. 3(b) and (c). We believe that dislocations in the austenite were also mobile during this experiment. The dislocation plasticity obscures the superelasticity which is caused by forward and reverse transformation occurring during loading and unloading.

Because the stress required for the onset of SIM ( $\sigma_M$ ) was not readily determined from the stress–strain curve, the external tensile stress required to form the first band of SIM was measured in several grains. The measurement was repeated and the results are shown in Fig. 4. After a SIM band was induced by loading, the specimen was heated under loading until the band disappeared (closed symbols). Then, the stress was increased again until the identical SIM band reappeared (open symbols) at a higher temperature. The two sets of symbols are fitted with two parallel straight lines, separated by less than 1 °C. The extrapolation of the line to the temperature axis gives  $A_f$  and  $M_s$ . Curves with different slopes were obtained in other

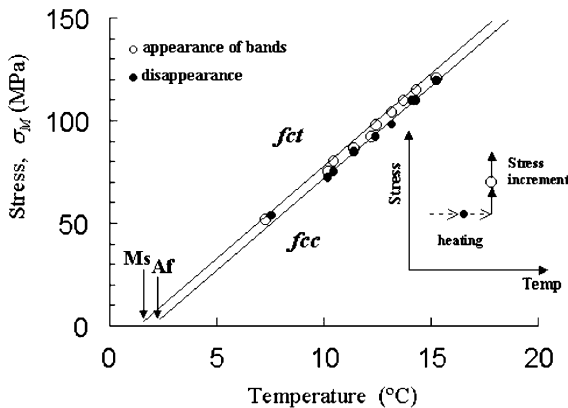


Fig. 4. Critical stress to induce martensite plates (open symbols) and the temperatures, at which these plates disappear by heating.

grains, possibly because the grains differed in orientation. The slope of the diagram shown in Fig. 4 is 4.8 MPa/°C, which is the smallest among the grains examined. This means that the transformation strain along the tensile direction of this grain was the largest.

Since the transformation was confirmed as first order, the tensile stress ( $\sigma_M$ ), transformation temperature ( $T$ ), transformation strain ( $\varepsilon$ ), and latent heat ( $H$ ) should satisfy the Clapeyron–Clausius relationship

$$d\sigma_M/dT = -\rho \cdot H/T\varepsilon, \quad (1)$$

where  $\rho = 8.91 \text{ g/cm}^3$  is the density of the FCC phase and  $H = -0.53 \text{ J/g}$  (DSC measurement). Using the minimum value of  $d\sigma_M/dT$ , the relationship gives the value of the axial strain as  $3.5 \times 10^{-3}$ . The transformation strain by a SIM plate is given below, using an infinitesimal approach [14]. The lattice parameter of the cubic austenite  $a_0$ , that of the  $c$ -axis in the tetragonal martensite  $c$  and that of the  $a$ -axis  $a$  are used to calculate the transformation strain. The transformation strain of a Bain correspondence variant which has the  $c$ -axis along the austenite  $[100]$  direction (BCV(1)) is written as

$$\varepsilon^T(1) = \begin{pmatrix} \varepsilon_c & 0 & 0 \\ 0 & \varepsilon_a & 0 \\ 0 & 0 & \varepsilon_a \end{pmatrix}. \quad (2)$$

Here,  $\varepsilon_c = (c - a_0)/a_0$ , and  $\varepsilon_a = (a - a_0)/a_0$ . The transformation strains of the other two variants are similarly written. There exist six types of twinned martensite plates. One of them has the average transformation strain

$$\bar{\varepsilon} = \begin{pmatrix} \varepsilon_a + \varepsilon_c & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \varepsilon_a \end{pmatrix}, \quad (3)$$

with  $\varepsilon_a > 0$  and  $\varepsilon_a + \varepsilon_c < 0$ . The average transformation strain of the other five plates is given by permuting the diagonal components in (3). If a tensile axis is in the standard triangle of  $[001]$ – $[011]$ – $[111]$ , the variant having the strain (3) causes the largest elongation,  $\varepsilon$ , along the tensile direction, given as,

$$\varepsilon = \varepsilon_a \cos^2 \theta + (\varepsilon_a + \varepsilon_c) \sin^2 \theta \cdot \cos^2 \phi. \quad (4)$$

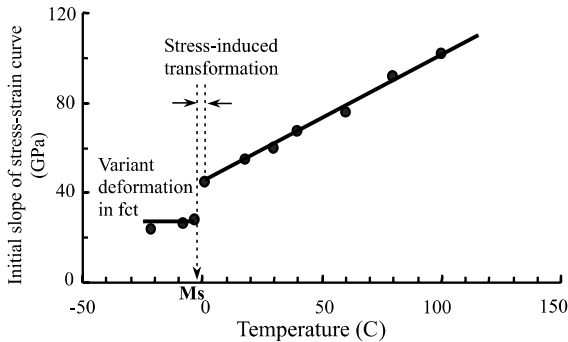


Fig. 5. Temperature dependence of initial slopes (up to 0.5% strain) in the stress–strain curve.

Here,  $\cos\theta$  is the direction cosine between the tensile direction and  $[001]$ , and  $\sin\theta \cdot \cos\phi$  that between the tensile direction and  $[100]$ . The elongation  $\varepsilon$  is largest ( $\varepsilon_a$ ), when the tensile axis is along  $[100]$  and smallest ( $(2\varepsilon_a + \varepsilon_c)/3$ ), along  $[111]$ , when the fraction of one Bain correspondence variant is  $\varepsilon_a/(\varepsilon_a - \varepsilon_c)$ . That is,  $\varepsilon$  ranges between  $\varepsilon_a$  and  $(2\varepsilon_a + \varepsilon_c)/3$ . Using the lattice parameters,  $a = 0.3790$  nm,  $c = 0.3695$  nm and  $a_0 = 0.3750$  nm from [10],  $\varepsilon_a$  and  $(2\varepsilon_a + \varepsilon_c)/3$  are estimated as  $1.1 \times 10^{-2}$  and  $2.2 \times 10^{-3}$ . The strain predicted from the Clapeyron–Clausius relationship falls in this range. To attain further agreement, a precise measurement of the lattice parameters, which are temperature dependent [9,10], is required.

Since strain due to stress-induced transformation contributes to the stress–strain relationship at temperatures close to  $M_s$ , the initial slopes (up to 0.5% strain) of stress–strain curves were measured as shown in Fig. 5. The slopes decreased gradually with decreasing temperature and showed a rapid decrease just above  $M_s$ . This decrease in slope is smaller than those reported previously [11,12]. Since our measurement used larger strain amplitude than the previous studies, which employed internal friction [11] and ultrasonic attenuation [12], the effect of the formation of SIM appears to overlap lattice softening just above  $M_s$ . Below  $M_s$ , variant change of FCT martensite predominantly occurs [9] and results in a smaller slope, shown in Fig. 5.

#### 4. Concluding remarks

At temperatures above  $A_f$ , the FCC/FCT transformation in an Fe–30.5 at.% Pd alloy is induced by external loading and the reverse transformation occurs during unloading. However, strain produced by loading does not completely recover by unloading. The initial slope (Young's modulus) of the stress–strain curve exhibits a sudden decrease, as Fe–Pd is cooled towards  $M_s$ . This is a consequence of lattice softening and is also caused by the occurrence of the stress-induced martensitic transformation.

#### Acknowledgements

We are grateful to Professor T. Mori for his stimulating discussion. Thanks are also due to Dr. M. Sugiyama of Nippon Steel Corporation and Dr. Katsushi Tanaka of Kyoto University for helpful advice on the present subject. This research was supported by New Energy and Industrial Technology Development Organization, Japan, and DARPA.

#### References

- [1] Wayman CM. MRS Bull 1993;49:49.
- [2] Chernenko VA, Cesari E, Kokorin VV, Vitenko IN. Scripta Mater 1995;33(8):1239–44.
- [3] Ullako K, Huang JK, Kantner C, O'Handley RC, Kokorin VV. Appl Phys Lett 1996;69:1966.
- [4] O'Handley RC. J Appl Phys 1998;83:3263–73.
- [5] Murray SJ, Farinelli M, Kartner C, Huang JK, Allen SM, O'Handley RC. J Appl Phys 1998;83:7297–9.
- [6] Tickle R, James RD. J Magn Magn Mater 1999;195: 627–38.
- [7] Bozhko AD, Vasil'ev AN, Khovailo VV, Buchel'nikov VD. J Exp Theor Phys 1999;8:954–62.
- [8] James RD, Wuttig M. Philos Mag A 1998;77:1273–99.
- [9] Sohmlura T, Oshima R, Fujita FE. Scripta Metall 1980;14:855–6.
- [10] Oshima R. Scripta Metall 1981;15:829–33.
- [11] Sugiyama M, Harada S, Oshima R. Scripta Metall 1985;19:315–7.
- [12] Muto S, Oshima R, Fujita FE. Acta Metall 1990;38:685–94.
- [13] Sugiyama M, Oshima R, Fujita FE. Trans JIM 1984;25(9):585–92.
- [14] Liang Y, Kato H, Taya M, Mori T. Scripta Mater 2000;43:535–50.