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The elastic modulus of B19' shape-memory NiTi was determined using three techniques; from the response of lattice planes measured using *in situ* neutron diffraction during loading, instrumented indentation using a spherical indenter and macroscopic extensometry. The macroscopic measurements resulted in a modulus of 68 GPa, significantly less than the 101 GPa from indentation and the lattice plane average of 109 GPa from neutron diffraction. Evidence from the neutron measurements suggests that the disparity derives from the onset of small amounts of twinning at stresses less that 40 MPa, which might otherwise be considered elastic from a macroscopic view point. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1863437]

In NiTi, a reversible thermoelastic martensitic transformation can be induced by temperature or stress between a cubic (B2) phase and a monoclinic (B19') phase.[1,2] Monoclinic NiTi (commonly called martensite) can twin to produce strains as large as 8%. Heating results in a phase transformation from the monoclinic phase to the cubic phase and associated recovery of all the accumulated strain, a phenomenon known as shape-memory. In addition to the commercial interest in the deformation behavior of shape-memory alloys for use as actuators, there is theoretical interest in probing deformation that involves twinning. The stress–strain deformation response of martensitic NiTi may include contributions from elastic, twinning and plastic deformation. In a macroscopic measurement using a strain gauge or an extensometer, it is difficult to distinguish between the different contributions. In particular, the relative contributions of twinning and plasticity are indistinguishable during loading. It is not unreasonable to expect that deformation at low stresses, largely expected to be elastic, includes twinning strains that result in an artificially low elastic modulus. While previous studies have suggested this,[3–5] direct evidence of twinning at low stresses in the context of elastic modulus measurements has not been observed thus far. By recourse to *in situ* neutron diffraction during loading, instrumented indentation and extensometry, the present work addresses these distinctions and obtains a modulus that is solely representative of elastic deformation.

The technique of using diffraction spectra for mechanical characterization relies on using atomic planes in specimens as internal strain gauges.[6] For the case of shape-memory alloys, performing such diffraction measurements *in situ*, i.e., by simultaneously recording diffraction spectra from samples that are subjected to external stresses or temperature changes, can provide bulk, quantitative, phase specific information on the evolution of strains, texture and phase volume fractions.[7–9]

The NiTi samples investigated in this work were fabricated using hot isostatic processing (HIP) with a nominal composition of 49.4 at % Ni, as previously described.[10] Since starting texture is known to influence the deformation behavior of shape-memory alloys, HIP was used to produce samples with a random distribution of grains. The lack of texture in NiTi samples fabricated using HIP has previously been confirmed.[10] Prior to testing, samples were subjected to a solutionizing treatment at 930 °C for 1 h under titanium gettered flowing argon and furnace cooled to room temperature.

Neutron diffraction measurements were performed in "time-of-flight" mode using the neutron powder diffractometer (NPD) at the pulsed neutron source at Los Alamos Neutron Science Center (LANSCE), Los Alamos National Laboratory (LANL). Samples were loaded in uniaxial compression and tension while neutron diffraction spectra were simultaneously collected. The cylindrical compression sample had a diameter of 10 mm and a gauge length of 12.5 mm while the tension sample had a rectangular cross section of 7×4 mm^2 and a gauge length of 25 mm. The load axis was placed horizontally at 45° to the incident neutron beam (5×10 mm pulsed / standard collimation) and a detector (at −90°) recorded diffraction spectra with scattering vectors parallel to the loading axis. A second detector at +90° was also used but that spectra is included in a subsequent analysis.[11] Neutron diffraction spectra were obtained at six compressive stresses (5, 33, 66, 100, 200, and 300 MPa) and nine tensile stresses (5, 33, 75, 125, 150, 180, 200, 220, and 240 MPa) during loading. At each stress, a spectrum was acquired for about 6 h at an average neutron beam current of 55 μA. The ramp rate was 0.1 mm/min. An extensometer was attached to the samples to record macroscopic strain during the experiments. The macroscopic stress–strain response determined from the extensometer during compres-

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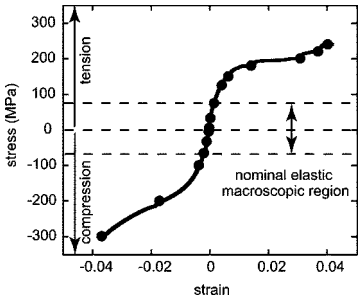

FIG. 1. Applied stress vs macroscopic strain measured by extensometry for NiTi in tension and compression. The symbols indicate the stresses at which neutron diffraction spectra were obtained during loading.

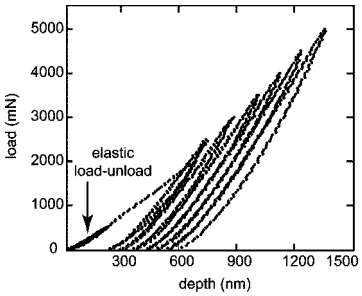

FIG. 3. Nanoindentation response of NiTi using a spherical diamond indenter. Load–unload experiments at the same location were performed to loads of 0.5, 2, 2.5, 3, 3.5, 4, 4.5, and 5 N. The initial elastic response corresponding to a load–unload experiment to 0.5 N is marked.

sive and tensile loading is shown in Fig. 1. For completeness, in addition to the initial elastic response, Fig. 1 includes regions where twinning is the dominant deformation mechanism that is analyzed in a subsequent publication.[11] A fit to the initial linear portion (up to 40 MPa) of the tension and compression stress–strain response in Fig. 1 resulted in a Young's modulus of 68±5 GPa (estimated error).

From the acquired neutron diffraction spectra, strains for specific grain orientations were determined by fitting individual lattice peaks with respect to the unloaded state using the RAWPLOT program in the General Structure Analysis System Software (GSAS).[12] A nominal holding stress of 5 MPa was used as the "zero stress" unloaded condition. The strain, ε_{hkl}, for a plane, *hkl*, at a given stress was calculated from

$$\varepsilon_{hkl} = \frac{d^{hkl} - d_0^{hkl}}{d_0^{hkl}}, \tag{1}$$

where d^{hkl} was the spacing of the plane at a given stress and d_0^{hkl} was the corresponding lattice spacing in the unloaded or "zero stress" condition. Since the strains are relative, the presence of pre-existing residual intergranular stresses was ignored. Figure 2 shows the stress–strain response of selected individual lattice plane reflections. From this figure, the elastic moduli during compressive and tensile loading were determined to be (values for tension in parentheses) 122 (113), 91 (92), and 108 (130) GPa for the 011, 100, and 111 planes, respectively. The estimated error in the elastic moduli values is ±5 GPa.

Instrumented indentation provides a record of the indentation load (P) as a function of the penetration depth (h) of a tip (usually a diamond) into a specimen. From the recorded load-depth or $P-h$ curve, various characteristic mechanical properties can be estimated.[13,14] Previous work has used indentation to qualitatively investigate shape-memory alloys.[15–18] In the following, a spherical diamond indenter was preferred over a sharp indenter in order to obtain a nonsingular stress field below the indenter and delay the onset of twinning and plasticity (in terms of indentation depth) during deformation.

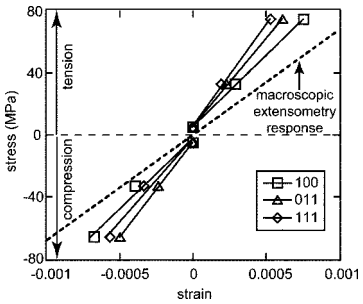

FIG. 2. The stress–strain response of individual lattice plane reflections in NiTi during loading in tension and compression.

A Nanotest 600 (Micromaterials Limited, Wrexham, UK) instrument was used to indent the aforementioned NiTi specimen. A custom spherical diamond indenter (2.8 mm diameter) was used and load-depth curves were obtained at a rate of 150 mN/s. The choice of this diameter was motivated by the need to indent about 10 grains for the depths reported. The machine compliance was determined by elastically indenting a standard steel specimen of know modulus. As an added check, the machine compliance determined using steel was used to determine the modulus of a fused quartz standard and values within 2 GPa of the standard were obtained. Each indentation experiment on the NiTi sample consisted of indenting the specimen to a load of 0.5 N and unloading. The absence of substantial residual depth on unloading was suggestive of the indentation response corresponding to mostly elastic deformation. As shown in Fig. 3, subsequent indents were performed at the same location as the 0.5 N indent, to loads of 2, 2.5, 3, 3.5, 4, 4.5, and 5 N, with complete unloading between indents at each depth. The purpose of adopting such a procedure was to progressively twin NiTi following the first predominantly elastic indent so that subsequent indents at the same location captured the response of the twinned martensite. Loading beyond 5 N was not pursued as there was no increase in residual indentation depth following unloading at greater loads. This implied that 5 N was adequate to obtain a fully twinned microstructure.

For elastic loading of a flat surface using a spherical indenter, the relationship between the load (P) and depth (h) is given by[19]

$$P = Ch^{3/2}, \tag{2}$$

where

$$C = \frac{2\sqrt{2}}{3} E^* D^{1/2}, \tag{3}$$

D is the indenter diameter and

$$E^* = \left(\frac{1-\nu_1^2}{E_1} + \frac{1-\nu_2^2}{E_2} \right)^{-1}. \tag{4}$$

E^* is a reduced elastic modulus that accounts for nonrigid indenters, where E_1 and ν_1 represent the elastic modulus and the Poisson's ratio of the indenter, respectively, and E_2 and

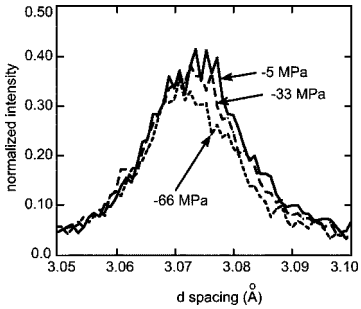

FIG. 4. Section of normalized neutron diffraction spectra from NiTi at −5, −33, and −66 MPa (compressive loading) corresponding to 011 lattice plane reflections.

ν_2 represent the elastic modulus and the Poisson's ratio of the material being indented, respectively. The elastic portion of the first (0.5 N) and last (5 N) indents were fit to a $3/2$ power law to determine C in Eq. (3). Using a value of 1141 GPa for diamond, 0.07 and 0.35 (Ref. 20) for the Poisson's ratios of diamond and NiTi, respectively, the elastic modulus of NiTi was determined to be 101 ± 7 GPa and 162 ± 6 GPa for the first (0.5 N) and last indents (5 N), respectively. The errors reported represent a standard deviation from 15 comparable indents.

The Young's modulus of martensitic NiTi determined from extensometry in this work (68 ± 5 GPa) is comparable with values previously reported and justified (69 ± 4 GPa).[20] Even values as low as 20–50 GPa have been reported.[4,21] The extensometry values are substantially lower than the values obtained here from neutron diffraction and indentation. Given the low symmetry of monoclinic NiTi, it is not straightforward to account for elastic anisotropy and obtain an average Young's modulus from the neutron diffraction measurements. However, the moduli of the lattice planes reported can be expected to reflect the average bulk polycrystalline response as seen previously for the case of cubic (B2) NiTi.[8] This is consistent with the modulus determined from indentation (101 GPa) lying in the range of lattice plane moduli determined from neutron diffraction (91–130 GPa). In the context of the aforementioned lower modulus from extensometry, Fig. 4 is presented to establish twinning at low stresses, in what is nominally an elastic regime for the macroscopic measurement. Figure 4 shows the 011 peak intensities with increasing compressive stress from neutron diffraction spectra that have been normalized so that spectra at each stress have the same total intensity (neutron counts). $(1\ 1\bar{1})$ Type I twinning in martensitic NiTi has been shown to result in 011 planes lying parallel to the loading axis under compressive loading.[20] From the scattering geometry, this would result in the 011 peaks decreasing in intensity, for the detector considered. This is indeed the case in Fig. 4, confirming that at very low stresses (33 and 66 MPa) martensitic NiTi undergoes twinning that results in additional strains, that in turn result in a lower Young's modulus from extensometry.

While the technique of neutron diffraction inherently measures elastic strain (as opposed to nonelastic strains which can only be measured when they influence elastic strains) and is not expected to be significantly influenced by the limited twining at low stresses, it is interesting to examine why instrumented indentation is not influenced by twinning as is extensometry. For this, consider that the in-plane shear stress under a spherical indenter is maximal at a ratio of depth to indenter contact half-width of 0.48, and not directly below the indenter.[22] The surrounding material at this depth may spatially constrain twinning at low stresses. Twinning eventually does occur, but at higher stresses as seen from the increase in modulus between the first and last indents (Fig. 3).

The present work suggests stress-strain curves of martensitic NiTi using macroscopic extensometry includes strain from twinning that results in a low Young's modulus, that are not truly representative of elastic deformation. Twinning at stresses as low as 33 MPa has been experimentally observed using neutron diffraction. On the other hand, determination of the elastic modulus using instrumented spherical indentation and lattice plane moduli from *in situ* neutron diffraction during loading result in values that are about 50% higher and are not influenced by twinning.

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[1] T. W. Duerig, K. N. Melton, D. Stoeckel, and C. M. Wayman, *Engineering Aspects of Shape Memory Alloys* (Butterworth–Heineman, Essex, 1990).
[2] *Shape Memory Materials*, edited by K. Otsuka and C. M. Wayman (Cambridge University Press, Cambridge, 1998).
[3] K. N. Melton and O. Mercier, Metall. Trans. A **9A**, 1487 (1978).
[4] Y. Liu and H. Xiang, J. Alloys Compd. **270**, 154 (1998).
[5] C. P. Frick, A. M. Ortega, J. Tyber, K. Gall, and H. J. Maier, Metall. Mater. Trans. A **35A**, 2013 (2004).
[6] *Measurement of Residual and Applied Stress using Neutron Diffraction*, NATO ASI Series E No. 216, edited by M. T. Hutchings and A. D. Krawitz (Kluwer Academic, Dordrecht, 1992).
[7] M. A. M. Bourke, R. Vaidyanathan, and D. C. Dunand, Appl. Phys. Lett. **69**, 2477 (1996).
[8] R. Vaidyanathan, M. A. M. Bourke, and D. C. Dunand, J. Appl. Phys. **86**, 3020 (1999).
[9] R. Vaidyanathan, M. A. M. Bourke, and D. C. Dunand, Acta Mater. **47**, 3353 (1999).
[10] R. Vaidyanathan, D. C. Dunand, and U. Ramamurty, Mater. Sci. Eng., A **289**, 208 (2000).
[11] C. R. Rathod, D. W. Brown, M. A. M. Bourke, and R. Vaidyanathan (unpublished).
[12] A. C. Larson and R. B. V. Dreele, General Structure Analysis System (GSAS). Los Alamos National Laboratory LAUR 8-748, (1986).
[13] S. Rajagopalan and R. Vaidyanathan, JOM **54**, 45 (2002).
[14] A. C. Fischer-Cripps, *Nanoindentation* (Springer-Verlag, New York, 2002).
[15] C. R. Rathod, S. Rajagopalan and R. Vaidyanathan, "Mechanical Characterization of Shape-Memory Alloys using Diffraction and Instrumented Indentation," Shape-Memory and Superelastic Technologies 2003, International Organization on Shape-Memory and Superelastic Technology, (2004), pp. 331–339.
[16] K. Gall, K. Juntunen, H. J. Maier, H. Sehitoglu, and Y. I. Chumlyakov, Acta Mater. **49**, 3205 (2001).
[17] W. Ni, Y.-T. Cheng, and D. S. Grummon, Surf. Coat. Technol. **177-178**, 512 (2004).
[18] N. Wangyang, Y. T. Cheng, and D. S. Grummon, Appl. Phys. Lett. **80**, 3310 (2002).
[19] J. Alcala, A. E. Giannakopoulos, and S. Suresh, J. Mater. Res. **13**, 1390 (1998).
[20] D. C. Dunand, D. Mari, M. A. M. Bourke, and J. A. Roberts, Metall. Mater. Trans. A **27**, 2820 (1996).
[21] D. E. Hodgson, M. H. Wu, and R. J. Biermann, *Metals Hand Book* (ASM International, Ohio, 1990), Vol. 2, p. 897.
[22] K. L. Johnson, *Contact Mechanics* (Cambridge University Press, Cambridge, 1985).