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The formation of stress-induced martensite in superelastic NiTi was studied by neutron diffraction during uniaxial compressive loading and unloading. The respective phase fractions were determined as a function of the applied stresses using a Rietveld refinement with a March–Dollase texture formulation. Before loading, the specimen was fully austenitic. At the highest applied stress of -625 MPa, about 90% of the austenitic phase had transformed to martensite, with a concomitant macroscopic strain of -2.8%. Upon unloading, all of the stress-induced martensite reverted to austenite and the totality of the macroscopic strain was recovered. The propensity for various austenitic crystallographic orientations to transform at different stresses was determined and qualitative observation of this incipient texture in the austenite and of the inherent texture in the nascent martensite are reported. © *1996 American Institute of Physics.* [S0003-6951(96)01843-8]

NiTi alloys with near-equiatomic composition exhibit near room temperature a reversible, thermoelastic transformation between a high-temperature, cubic (B2) austenitic phase and a low-temperature, monoclinic (B19) martensitic phase.[1] Austenitic NiTi alloys with nickel-rich composition can show superelastic (or pseudoelastic) behavior: during mechanical loading, tensile strains as high as 8% result from the formation of stress-induced martensite; during unloading, the martensite becomes unstable and reverts to austenite, with concomitant recovery of all the accumulated macroscopic strain.[2–4] In related behavior, martensitic alloys exhibit a shape memory effect, for which deformation takes place by twinning of the martensite, and strain recovery occurs by a thermally-induced, rather than stress-induced, transformation.

In NiTi single crystals, surface observations have recorded the growth and disappearance of stress-induced martensite during a mechanical cycle.[5–7] Unfortunately surface studies are impractical for small grain polycrystalline samples. Alternatively, x-ray diffraction on polycrystalline NiTi wires has identified stress-induced changes in crystal structure.[8] However, x-ray penetration is limited to typically less than 40 μm below the surface where the state of stress and thus the transformation in superelastic materials are unrepresentative of the bulk. By contrast, neutron diffraction is ideally suited for the study of stress-induced transformations, because the average behavior of bulk polycrystals can be measured with sampling volumes of up to 1 cm^3. Although the irradiated volume is large compared to a typical polycrystalline microstructure, each diffraction peak is an average over many grains which have an orientation defined by the scattering geometry. Thus, while the transformation of an individual grain cannot be measured, the average behavior of many grains can be. At a pulsed source operated in time of flight mode, collection of an entire diffraction pattern occurs for each measurement, and, for a given detector, the scattering vectors of all reflections lie in the same direction. Thus, in a single measurement, the behavior of all crystallographic planes can be explored by aligning the load axis relative to the scattering vector

While neutron diffraction studies of twinning have been reported for martensitic shape-memory NiTi,[9–11] we are aware of only one neutron study of superelasticity,[12] whereby the structure of a Cu–Al–Ni single crystal was determined as a function of stress. In this letter, we present neutron diffraction results on polycrystalline superelastic NiTi subjected to a series of stress levels. We report quantitative measurements of the phase fractions and qualitatively discuss the texture evolution; the crystallographic elastic lattice strains will be discussed elsewhere.

A NiTi billet was fabricated by hot-isostatic pressing (1065 °C, 100 MPa, 3 h) of prealloyed powders blended with small amounts of pure nickel.[11] One cylindrical compression specimen (10 mm in diameter and 24 mm in length) was fabricated by electrodischarge machining, solutionized at 1000 °C for 1 h in argon and oil quenched to room temperature. Subsequently, it was annealed at 400 °C in air for 1 h and quenched in icewater. Prior to the diffraction analysis the specimen, outfitted with an extensometer, was subjected to two training (load–unloading) cycles with peak stresses of -563 and -550 MPa. A nonrecoverable plastic strain of -0.1% was recorded after the first training cycle but none was noted during the second or during the diffraction cycle, despite an increase in the maximum stress to -625 MPa.

Diffraction measurements were performed using the Neutron Powder Diffractometer at the Manuel Lujan, Jr. Neutron Scattering Center. The load was ramped in stroke control and kept constant during hold periods where diffraction spectra were collected. The ramp and hold periods were respectively, about 1 min and 2–4 h, depending on beam intensity. The load axis was placed in a horizontal plane, at 45° to the incident neutron beam, and two detectors recorded diffraction patterns with scattering vectors parallel and perpendicular to the load axis.[13,14]
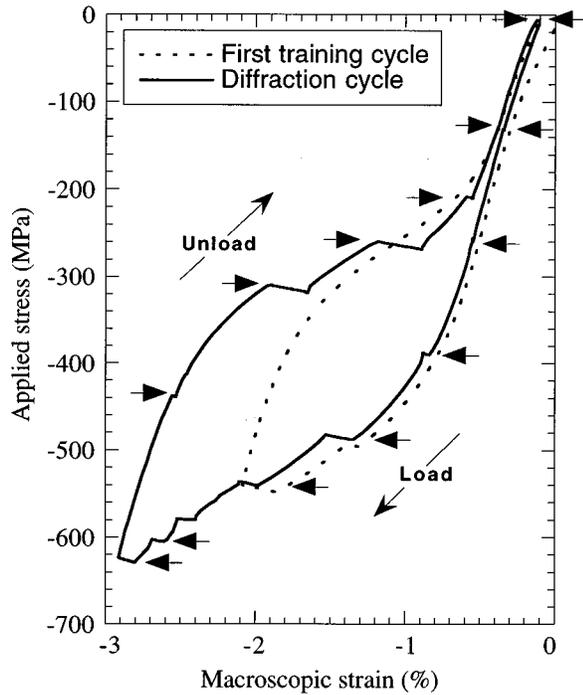
FIG. 1. Macroscopic stress-strain curve of superelastic NiTi for the first (training) and the third (diffraction) cycles. Stress levels where diffraction spectra were recorded are marked with arrows.

Diffraction spectra were recorded at 14 stress levels, as depicted in Fig. 1 which shows the stress-strain curve for the first training cycle and the third diffraction cycle. The stresses at which the diffraction measurements were made are apparent from the steps in Fig. 1, associated with small, time-dependent strains. These steps probably result from the sample temperature (increased or decreased by the transformation enthalpy) returning to room temperature during the hold period.

After yield at a stress of about −350 MPa, plastic defor-

FIG. 3. Section of diffraction spectra for all applied stresses. The spectra are smoothed, normalized, and displaced along the Y axis for clarity. The scattering vector is parallel to the loading direction.

mation results from the austenite-to-martensite transformation (Fig. 1). The difference in structure between the unstressed and stressed states are shown in Fig. 2 in which the broad martensitic peaks are clearly discernible from the sharp austenite peaks. The progression and reversibility of the stress-induced transformation are illustrated in Fig. 3, which shows a short section of the spectra for each stress level. Upon loading, the shift to shorter d spacings result from increasing elastic compressive strains, while the de-
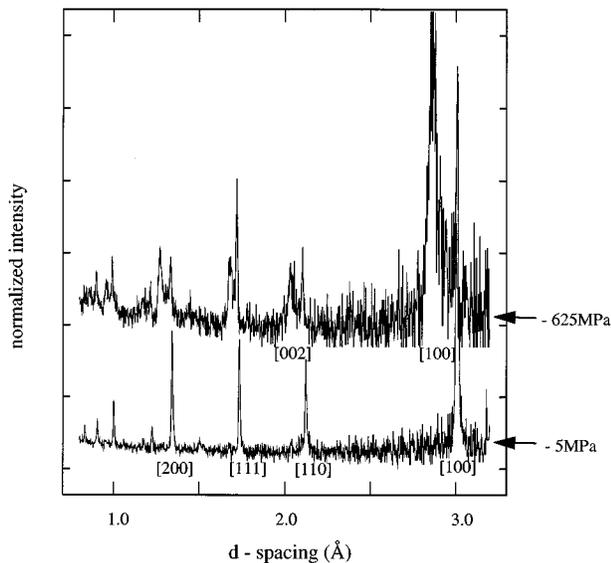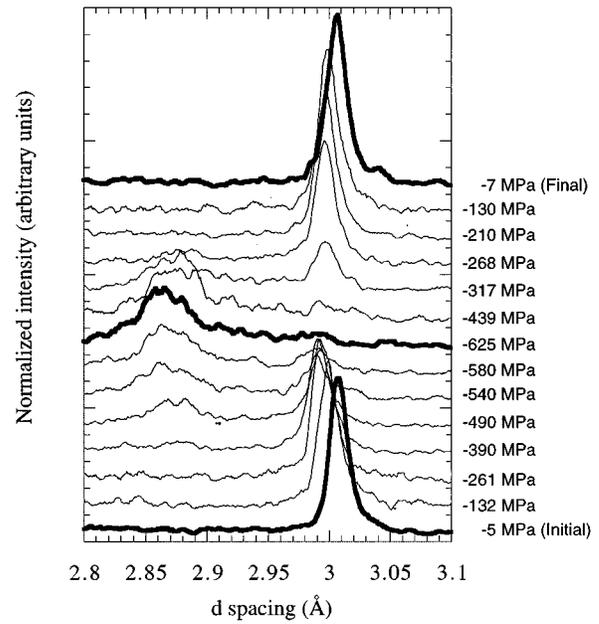

FIG. 2. Diffraction spectra for 100% austenite and 90% martensitic compositions (−5 and −625 MPa, respectively) with main peaks labeled. The scattering vector is parallel to the loading direction.
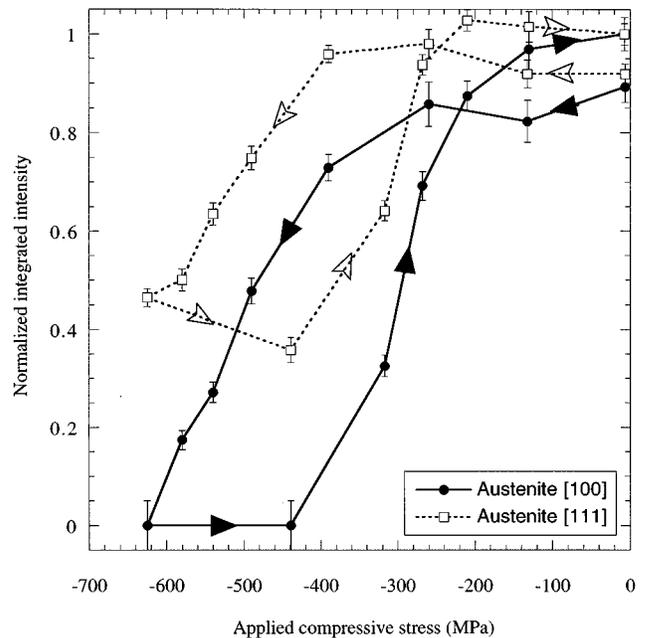

FIG. 4. Stress dependence of the intensity of [100] and [111] austenitic peaks normalized to the value after unloading. The scattering vector is parallel to the loading direction.

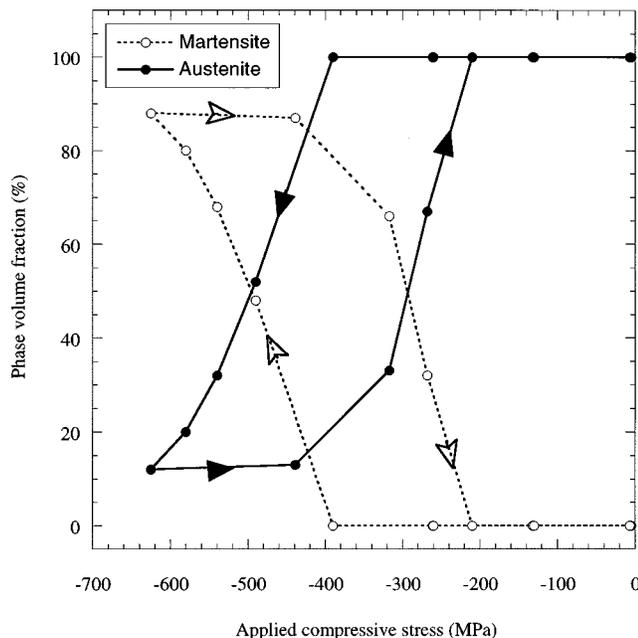   Bourke, Vaidyanathan, and Dunand

FIG. 5. Stress dependence of the volume fractions of the martensite and austenite phases through the load cycle.

crease in intensity of the austenitic peak and concomitant increase in the martensitic peak are due to transformation. As illustrated in Fig. 4, where the intensity of the austenitic [100] and [111] peaks are plotted as a function of stress through the loading cycle, not all austenitic reflections diminish at the same rate. At the maximum stress, the [100] intensity is close to zero whereas the [111] intensity is approximately half of the zero-stress value. The propensity for transformation is thus related to the crystallographic orientation of the austenite. One unusual observation was that the integrated intensity of the austenitic reflections in the unstressed state showed a small increase after the loading cycle but the effect has not been studied in detail.

The incipient texture in the austenite described above, resulting from the transformation of grains in preferred orientations prior to those in less favorable orientations, is matched by the strong orientation in the nascent martensite. For instance, the martensitic [010] peak was absent in spectra for which the scattering vector was parallel to the load, whereas the martensitic [100] peak was absent in spectra for which the scattering vector was perpendicular to the load. Rietveld refinements of the spectra (scattering vector parallel to load) were performed with a March–Dollase description of the texture using the Los Alamos Generalized Structure

Analysis System GSAS program.[15] The stress-dependence of the phase volume fractions determined by these refinements are shown in Fig. 5. An unexpected result is that about 90% of the austenite is transformed to martensite at a macroscopic strain of only -2.9%. Since the maximum recoverable strain in tension is 8%,[2–4] and since the superelastic effect for single crystals is similar in compression and in tension,[16] this raises the question of whether, at strains above -2.9%, further recoverable strain can be produced by twinning of the stress-induced martensite. An alternate explanation is that the total superelastic strain is lower in compression than in tension in polycrystalline superelastic NiTi specimens.

In summary, the significance of the present work lies in the potential to quantify in polycrystals the proclivity for transformation of specific grain orientations at different stresses and at different angles to the loading direction. The results also offer the potential to validate polycrystalline models and a mechanism to examine whether and how elastic anisotropy biases the transformation paths.

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