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Ultrahigh-Resolution *In Situ* Diffraction Characterization of the Local Mechanics at a Growing Crack Tip in Nitinol

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Abstract

Despite the recognized importance of the *in vivo* fatigue properties of Nitinol, there is still limited understanding of how fatigue cracks propagate in this material. This study represents an initial approach to gain such understanding via a combination of fracture-mechanics testing and synchrotron x-ray (micro) diffraction, by providing insight into the role of transformational and local strain fields on the progression of fracture in a stent-like material structure. The results presented here compare the fracture-mechanics predicted transformation-zone size and shape with the actual zones measured by micro-diffraction. Tests were conducted with compact-tension specimens, laser-cut from Nitinol tube that was shape-set flat; this configuration mimics the microstructure and texture observed in Nitinol medical devices. Fatigue cracks were grown *ex situ* at near-threshold conditions (ΔK = 3 MPa√m) to a crack length to sample width ratio of a/W = 0.5. Specimens were then loaded *in situ* with a miniature straining rig to various stress intensities for multiple fatigue cycles. Thousands of local diffraction patterns (1 μm^2 spot area) spanning hundreds of micrometers surrounding the crack tips, were combined to produce contour maps of phase volume and local strain. The differences in monotonic and cyclic loading conditions can be deduced from these tests and can be used to differentiate *in vivo* single-event versus cumulative-damage fractures.

Introduction

Few studies have addressed the problem of growing fatigue cracks in the Nitinol material used for biomedical devices. Most experimental work performed to date in this area [1-5] has been concerned with quantification of crack-growth parameters (i.e., Paris-law constants, fatigue thresholds, etc.), but not with the local stresses, strains and phase volumes ahead of a growing sharp crack. However, Labossiere and Perry [6] used Moiré interferometry to reveal local strains and visually captured the surface relief ahead of the notch tip that was presumed to be associated with the austenite-to-martensite phase transformation. Unfortunately, these results were conducted at a notch tip rather than an atomically sharp fatigue crack, and were performed on sheet material that is not relevant to the manufacture of endovascular stents. Furthermore, surface relief as a means for measuring phase transformation may be misleading. Additionally, McKelvey and Ritchie [3] used transmission electron microscopy (TEM) to conclude that the phase transformation ahead of a crack tip could be suppressed by constraint in thick-section compact-tension specimens. Corresponding numerical simulations have also been performed [6-

9]; these purport to estimate the local strains, stresses and phases ahead of a crack, but are limited by somewhat questionable constitutive models employed in the finite element analyses.

It is the goal of the present research to investigate how the *in situ* phase transformation affects the distribution of strains ahead of a growing (nominally atomically-sharp) crack in Nitinol used for biomedical device manufacture. Using a combination and fracture mechanics and synchrotron x-ray diffraction, our approach is to load miniaturized compact-tension specimens of Nitinol flattened tubes in tension and to examine the growing crack tip *in situ* with the high spatial resolution of synchrotron radiation x-ray micro-diffraction beamlines. This technique allows the measurement of highly localized strains (with 1 μm resolution) and phase distributions by diffraction.

Experimental Procedures

Material

Nitinol samples with a composition of 50.8 atomic percent Ni, in the form of compact-tension C(T) specimens, were received from Nitinol Devices & Components, Inc. (Fremont, CA). Nitinol tubing, similar to that used for manufacture of stents, was cut longitudinally, then unrolled and shape-set flat. The heat treatments required to produce flattened material from the original tube configuration was similar to a typical shape-setting procedure utilized for stent manufacture, both in annealing time and temperature. Additional annealing at 735°C for 3-5 min was performed to yield an average grain size of 5-10 μm; this annealing step to increase the grain size was necessary for expeditious diffraction pattern collection. The flattened material was laser machined into C(T) specimens, with an overall size of 12 mm square and a thickness of 0.37-0.41 mm. Each sample was electropolished prior to testing to minimize surface discontinuities, and had an A_f temperature between 15º and 20°C. Specimens were machined such that the crack-growth direction was oriented at 45° to the tube-drawing direction, as previous studies had shown that fatigue-crack growth favored this direction regardless of the initial orientation of the pre-crack [5].

Experimental Setup

All samples were fatigue pre-cracked in ambient air to a crack length to width ratio of $a/W \sim 0.5$ using an MTS servo-hydraulic mechanical testing system, operating at a near-threshold ΔK of ~3 MPa√m, with a frequency of 50 Hz and a load ratio of 0.1. Samples were then loaded using a miniature tensile stage specially designed for use on the Advanced Light Source (ALS) microdiffraction beamline. The C(T) specimens were tested in displacement control using a Newport stepper motor, at 0.25 μm sec^{-1}, using pin grips; the resultant load output was measured with an Interface 450 N miniature load cell. Microdiffraction experiments were conducted at the ALS beamline 7.3.3 and utilized a diffraction spot size of ~1 μm^2, an energy range of 6-12 keV (white light), with a flux of 1.9 GeV at 400 mA. Thousands of Laue diffraction patterns were collected using a CCD camera in the region immediately surrounding the crack tip, with resolved areas ranging from 100 x 100 μm to 1 x 1 mm, with step sizes from 2-25 μm, and spot size always at ~1 μm^2. Laue diffraction patterns were indexed using XMAS software routine which enabled the computation of the local strain and phase maps around the crack tip.

Results and Discussion

Strains and Phase Distribution at a Crack Tip

The present study has utilized *in situ* x-ray microdiffraction to examine the nature of the phase transformation ahead of a growing crack tip in a material that is appropriate for endovascular

stents, specifically thin-walled superelastic Nitinol tubing; the approach includes the definitive identification of phases present while the test specimen is under load. Figure 1 shows that in such thin-walled Nitinol tubing, there is clearly evidence of a phase transformation ahead of the crack tip. The transformation-zone size at an applied load of 60N, corresponding to a mode I stress intensity of 15 MPa√m, is peanut-shaped and ~100 μm in width (i.e. parallel to the loading axis). This zone is identified by material that is 100% martensite within the entire x-ray diffraction spot region (1 μm square, with depth of penetration ~5 μm). We presume that there exists another intermediate zone with a mixed-phase structure; however, given the limitations of the current analysis, this intermediate zone cannot be resolved from the surrounding bulk untransformed material.

By applying standard linear-elastic fracture mechanics with a Mises yield criterion equal to the transformational stress (370 MPa), which clearly is not appropriate for superelastic material, or even modified yield surface specific to Nitinol [10], one determines that the zone of transformation should be ~600 μm, some six times larger than the size of the experimentally observed zone. However, this calculated zone size would include fully transformed plus mixed-phase material, in contrast to the experimental measurements which only reveal the fully transformed zone. To produce a fully transformed-zone size, which is experimentally determined to be ~100 μm, the Mises and Lexcellent & Blanc yield surfaces need to be increased to the yield stress of martensite (1000 MPa). The fact that a 1000 MPa yield surface produces the appropriate fully transformed-zone size suggests that many variables unique to Nitinol, other than just the tension-compression asymmetry, can affect the size/shape of the transformation zone. As will be discussed later, finite element codes that take into account a stress plateau in addition to many other variables not accounted for by yield surface analyses do accurately predict the transformation size/shape at a much lower stress value.

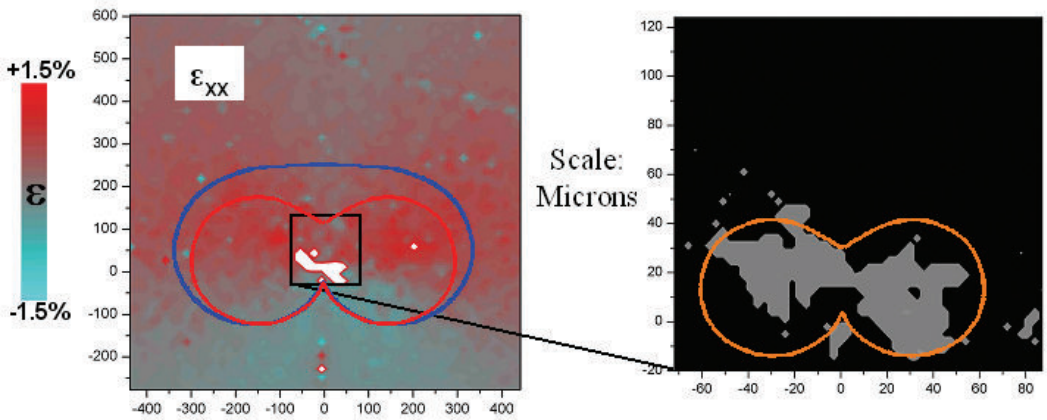

Figure 1: Strain and phase distributions shown in a C(T) sample loaded to 60N (stress intensity 15 MPa√m); peanut-shaped white region in left figure and gray region in right figure show the experimentally measured fully transformed material zone. The blue and red zones boundaries in the left diagram show respectively the theoretical transformation zones (which include fully, and partially, transformed material) for Mises and Lexcellent & Blanc [10] yield surfaces assuming a 370 MPa transformational stress. The orange zone boundary in the right figure represents the theoretical zone size utilizing Lexcellent & Blanc's Nitinol yield surface with a yield stress equivalent to martensite yielding (1000 MPa) rather than the plateau stress (370 MPa). Crack growth direction is vertical from bottom to top.

The shape of the transformed zone is counterintuitive if one assumes standard linear-elastic fracture mechanics. Since the x-ray penetration depth is only 5 □m and the sample thickness is ~400 □m, the probed area should be in a state of plane stress. However, a peanut-shaped zone typical of a plane-strain condition is observed ahead of the crack tip. If one takes into account the tension-compression asymmetry, as Lexcellent & Blanc [10] did in creating a Nitinol yield surface, a peanut-shaped zone is produced even under plane-stress conditions. It is therefore critical when modeling transformation zones in Nitinol that the many characteristic properties of this material are considered, specifically the tension-compression-torsion asymmetry, the modulus mismatch, the temperature above the A_f, and the crystallographic texture.

Evolution of Martensite with Fatigue Loading

A recent study of strain-controlled cyclic fatigue showed a decay of the modulus with increasing number of fatigue cycles. This behavior is likely due to pinning of dislocations and non-reversible martensite [11]. Therefore, in the present study, the evolution of the transformation zone was studied by x-ray microdiffraction to monitor the change in size and shape of the zone with increasing cycles. The C(T) specimens were loaded between ~30 to 60 N for 0 to 100 cycles, corresponding respectively to stress intensities 7.5 to 15 MPa√m (at a load ratio of ~0.5). Loading to a maximum value (at 15 MPa√m) produced a fully transformed zone of ~150 μm in width, which was stable both in size and shape throughout the entire loading process. Conversely, initial loading at cycle 0 up to 7.5 MPa√m, produced an extremely small, almost undetectable, fully transformed zone; this fully transformed zone size increased in size with increasing number of cycles, until it finally stabilized after 2 to 10 loading cycles. Interestingly, despite being well within the transformation zone, the same grains (orange and yellow grains in the left side of the transformed zone in Fig. 2) were resistant to transformation at both the lower and upper loads applied. The resistance to transformation of similarly oriented grains suggests that crystallographic orientation (texture) plays an important role in the phase transformation, even under sharp strain-gradient conditions.

Comparison between Experimental and Numerical Results

One goal of this work was to verify the ability of finite element modeling (FEM), specifically the ABAQUS Elastic-Superelastic Nitinol Subroutine (v. 6.5-4), to accurately predict transformation zone sizes and shapes in a complex mechanical geometry e.g., C(T) fracture specimens. With such crystallographic-mechanics information, these codes can more accurately predict fatigue behavior in more complex geometries such as stents. The ABAQUS subroutine takes into account many properties associated with the deformation and fracture behavior that makes Nitinol unique: stress plateau, elastic modulus mismatch between austenite and martensite, hysteresis loop, superelasticity, and tension-compression anisotropy. Three zones are predicted from the FEM calculations, namely those associated with untransformed austenite, partially transformed material, and fully martensite region immediately surrounding the crack tip. Figure 3 shows that the predicted fully transformed zone (indicated in red) is peanut-shaped and ~100 μm in width. This numerical prediction correlates extremely well with the experimentally-measured fully transformed zone size and shape, shown in Fig. 1.

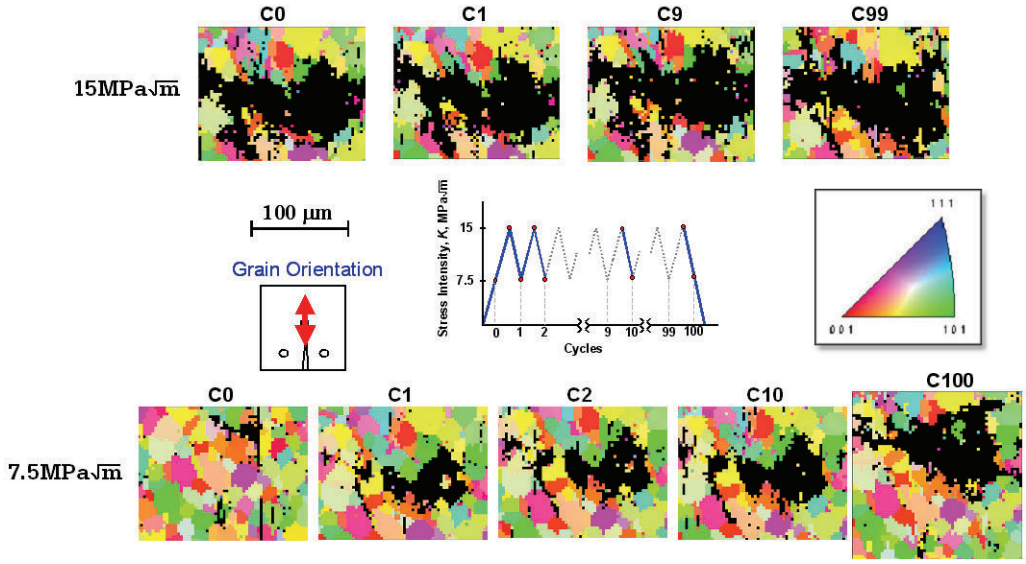

Figure 2: Evolution of a fully transformed martensite zone ahead of a crack tip following multiple fatigue cycles, showing stable zone size at maximum load, and a growing zone size at minimum load that stabilized between cycle 2 and 10. Grain orientations with respect to the crack-growth direction are shown via a color grid with red, green and blue corresponding to 100, 110, and 111 crystalline orientations, respectively. Crack growth is vertical from bottom to top in each grain map.

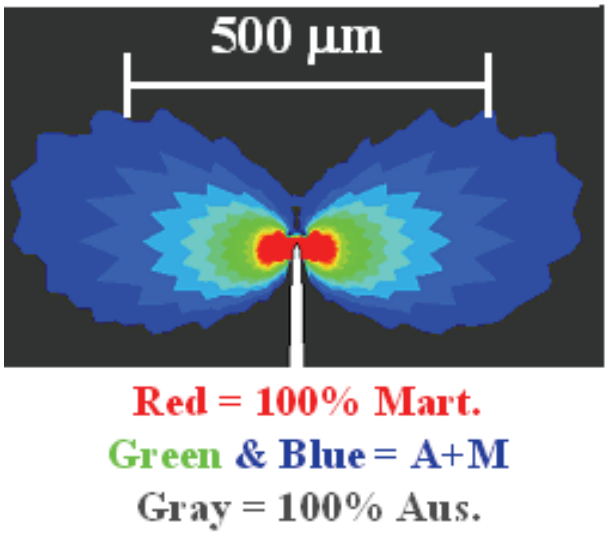

Figure 3: Simulated transformation zone using ABAQUS Nitinol Subroutine (v. 6.5-4) showing three primary zones: fully austenite (gray), mixed phase (green and blue), and fully martensite (red). The finite element model predicts that the fully transformed material (red) is peanut shaped and ~100 µm in width (applied loading direction), and thus corresponds extremely well with the experimental data.

Conclusions

We have presented initial results in which we combine standard fracture-mechanics experiments with high-spatial-resolution synchrotron radiation X-ray microdiffraction. Our goal is to measure the *in situ* strain and phase distributions surrounding a growing fatigue crack in Nitinol tube. Due to current limitations of the analysis, mixed phase regions were identified as strained austenite, such that only the fully transformed material zone was imaged by the experimental analysis. When a ~60N load was applied to the samples, corresponding to stress intensity of 15 MPa√m, the fully transformed zone was ~100 μm in width and was peanut shaped. This zone shape could not be predicted by standard linear-elastic fracture mechanics. Instead, more complex analyses that incorporated the elastic modulus mismatch between phases, hysteresis loop behavior, and the tension-compression asymmetry of Nitinol were required to accurately predict the peanut-shaped transformation zone ahead of the crack tip. Upon cyclic loading, the transformation zone stabilized immediately at the maximum load, but progressively grew in size at the minimum load, stabilizing only after 2 to 10 fatigue cycles. Furthermore, similarly oriented grains that were clearly within the transformation zone appeared to suppress the transformation, suggesting that texture plays an important role in the driving force to form martensite from austenite.

Acknowledgements

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