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Elastic properties of rolled uranium–10 wt.% molybdenum nuclear fuel foils

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In situ neutron diffraction data was collected during elastic loading of rolled foils of uranium–10 wt.% molybdenum bonded to a thin layer of zirconium. Lattice parameters were ascertained from the diffraction patterns to determine the elastic strain and, subsequently, the elastic moduli and Poisson's ratio in the rolling and transverse directions. The foil was found to be elastically isotropic in the rolling plane with an effective modulus of 86 ± 3 GPa and a Poisson's ratio 0.39 \pm 0.04. - 2013 Published by Elsevier Ltd. on behalf of Acta Materialia Inc.

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In support of the United States' nonproliferation and highly enriched uranium (HEU) minimization policies, the US Department of Energy (DOE)/National Nuclear Security Administration's Global Threat Reduction Initiative (GTRI) is actively working to convert civilian research and test reactors from the use of HEU fuel to low enriched uranium (LEU) fuel [\[1\]](#page--1-0). GTRI's Reactor Conversion program provides governments and facilities around the world with technical and economic assistance for conversion. If no suitable LEU fuels are available, the program contributes to the development of new LEU fuels. To date, GTRI has converted or verified the shutdown of 87 research reactors worldwide, including 20 domestic facilities. Of the remaining domestic research reactors, five US high-performance research reactors and one associated critical assembly will require a new high-density LEU fuel and fabrication capability, which is currently under development, to convert.

Existing qualified fuels do not meet the high fuel density requirements for the operation of these high-performance reactors, which include the Advanced Test Reactor at Idaho National Laboratory, the High Flux Isotope Reactor at Oak Ridge National Laboratory, the University of Missouri Research Reactor, the Massachusetts Institute of Technology Reactor and the Department of Commerce's National Bureau of Standards Reactor. To maintain performance requirements, the Reactor Conversion program is developing a high density monolithic plate fuel system which uses LEU–10 wt.% molybdenum (U-10Mo) foils clad with Al [\[2\].](#page--1-0) A thin $(\sim 0.025 \text{ mm})$ zirconium inter-diffusion barrier layer on the U-10Mo foil prevents the formation of U–Al intermetallics, the properties of which may be detrimental to the fuel performance [\[1–3\]](#page--1-0).

The fuel and the cladding are bonded using hot isostatic pressing (HIP'ing). Due to the mismatch between the coefficients of thermal expansion of the constituents, significant residual stresses must develop between the Al and U-10Mo components during cooling from the HIP'ing temperature of 560 \degree C. We have initiated diffraction measurements of residual stress in monolithic Al-clad U–10Mo fuel plates and others [\[4,5\]](#page--1-0) have utilized finite element analysis to model the evolution of stresses, but the literature lacks definitive elastic moduli data for the heavily rolled, strongly textured material necessary for both the experimental and modeling analysis.

There is considerable variation in the literature for the elastic properties of U-10Mo. The reported elastic moduli in bulk and relatively weakly textured samples [\[6–9\]](#page--1-0) are in the range of 84–92 GPa. The value of 65 GPa [\[10,11\],](#page--1-0) which was observed on a thin foil

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(0.250 mm) that had been heavily rolled and had a strong texture, raises the possibility of a very anisotropic elastic response. None of the previous work has explicitly addressed the potential anisotropy of the elastic modulus in the body-centered cubic (bcc) UMo alloys with strong texture. For comparison with more familiar bcc materials, we note that textured ferritic steel has an anisotropic elastic modulus while the elastic properties of tungsten are isotropic [\[12\].](#page--1-0)

To address this issue, neutron diffraction measurements were completed in situ during elastic loading of heavily worked thin foil U-10Mo specimens, including the Zr inter-diffusion layer. Neutron (and X-ray) diffraction are unique in that they are non-contact techniques to measure the strain. Moreover, diffraction unambiguously measures only the elastic strain and separates the response of multiple phases in what is essentially a composite sample.

The Zr-bonded U-10Mo foils were fabricated at Los Alamos National Laboratory (LANL). The U-10Mo alloy was vacuum-induction melt cast into a book mold with dimensions of approximately 150 mm \times 200 mm \times 1.5 mm. The as-cast piece was hot-rolled to \sim 2.5 mm thickness, being held at 670 °C in a salt bath between passes. This piece was sectioned into a coupon, cleaned and further co-rolled with the Zr inter-diffusion layers in a sealed steel can to 0.5 mm (U-10Mo + Zr) thickness. For co-rolling, the foil was heated in an air furnace to $680 °C$ prior to each pass. After the rolling was complete, the canned material was held in the air furnace at $680 \degree$ C for half an hour before allowing it to cool to room temperature. The Zr-bonded U-10Mo was then removed from the steel rolling can and coldrolled from 0.5 mm to the final dimension of nominally 0.33 mm in seven passes with no final anneal performed. Dogbone-shaped tensile samples with a gauge length of 17 mm and a total length of 50 mm were electric discharge machined from the foil with tensile axis parallel to the rolling (RD) and transverse (TD) directions. A single sample from each in-plane sample direction was tested to determine the elastic moduli.

The in situ neutron diffraction measurements of the lattice parameter during elastic loading were performed on the Spectrometer for Materials Research at Temperature and Stress (SMARTS) at the Lujan Neutron Scattering Center, LANSCE, LANL. Details of the diffraction instruments are published elsewhere [\[13\],](#page--1-0) so only a short description is presented here. SMARTS has a 30 m primary flight path and 1.5 m secondary flight path, making it suitable to measure the

small changes in lattice spacing associated with elastic strains. A purpose-built horizontal load frame (factory calibrated annually) was utilized to perform in situ measurements of the lattice strains during uniaxial tension. The load axis was oriented at 45° relative to the incident beam. Detectors on either side of the specimen simultaneously record data with diffraction vectors, parallel Q_{\parallel} (-90°) and transverse Q_+ (+90°), to the applied load [\[14\].](#page--1-0) The diffractometer was calibrated using a standard calcium fluoride powder sample in a vanadium container.

Each diffraction peak (hkl) comprises neutrons diffracted from a unique subset of grains from within the irradiated volume with a common orientation, that is, a specific crystallographic plane normal (hk) that is parallel to the diffraction vector defined by the instrument geometry [\[15\]](#page--1-0). Thus, the response of a subset of grains with a specific orientation relative to the loading direction is indicated by the evolution of the associated diffraction peak. The effective elastic properties of that specific grain family, which are dependent on the bulk crystallographic texture through the interaction of the grain set with its average neighborhood, are then found from the evolution of the interatomic $(d₋)$ spacing as a function of load in the elastic regime. In contrast, the lattice parameter of each phase, determined by Rietveld refinement, effectively averages over all grain orientations and represents the average phase response [\[16\].](#page--1-0) Then, in either case, the evolution of the lattice parameter or d-spacing determined from the diffraction patterns collected in the Q_{\parallel} and Q_{\perp} detector banks during elastic loading provides the effective elastic modulus and Poisson's ratio, respectively.

Figure 1 shows a schematic and picture of a tensile sample and the grips, which were friction based. A screw passed through two knurled plates on either side of the sample and through the hole in each end of the sample. A secondary plate with thickness matching the sample thickness kept the knurled plates parallel as the screw was tightened on the knurled plates, providing a large normal, and thus frictional, force on the sample. Tensile force was then applied by a hydraulic actuator via a pin through the secondary plate on one side of the sample and measured by a 10 kN load cell, again via a pin, through the secondary plate on the other side of the sample.

The stress was increased incrementally under load control to a maximum level of 300 MPa, still in the elastic regime. After each increment, neutron diffraction data was collected for 30 min in order to determine the

Figure 1. Schematic and picture of tensile sample in grips.

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