***In Situ* Synchrotron Tensile Investigations on Ultrasonic Additive Manufactured (UAM) Zirconium**

Chemical and Fuel Cycle Technologies Division

About Argonne National Laboratory

Argonne is a U.S. Department of Energy laboratory managed by UChicago Argonne, LLC under contract DE-AC02-06CH11357. The Laboratory's main facility is outside Chicago, at 9700 South Cass Avenue, Lemont, Illinois 60439. For information about Argonne and its pioneering science and technology programs, see www.anl.gov.

DOCUMENT AVAILABILITY

Online Access: U.S. Department of Energy (DOE) reports produced after 1991 and a growing number of pre-1991 documents are available free at OSTI.GOV (<http://www.osti.gov/>), a service of the U.S. Dept. of Energy's Office of Scientific and Technical Information.

Reports not in digital format may be purchased by the public from the National Technical Information Service (NTIS):

U.S. Department of Commerce
National Technical Information Service
5301 Shawnee Rd
Alexandria, VA 22312
www.ntis.gov
Phone: (800) 553-NTIS (6847) or (703)
605-6000 Fax: (703) 605-6900
Email: **orders@ntis.gov**

Reports not in digital format are available to DOE and DOE contractors from the Office of Scientific and Technical Information (OSTI):

U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831-0062
www.osti.gov
Phone: (865) 576-8401
Fax: (865) 576-5728
Email: **reports@osti.gov**
Disclaimer

Disclaimer

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor UChicago Argonne, LLC, nor any of their employees or officers, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of document authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof, Argonne National Laboratory, or UChicago Argonne, LLC.

***In Situ* Synchrotron Tensile Investigations on Ultrasounic Additive Manufactured (UAM) Zirconium**

prepared by

Yinbin Miao¹, Kun Mo¹, Jun-Sang Park¹, Jonathan Almer¹, Caleb Massey², Cody Havrilak², Andrew T. Nelson², Heather Connaway¹, Abdellatif M. Yacout¹

1) Argonne National Laboratory

2) Oak Ridge National Laboratory

June 2021

(This page left intentionally blank)

Abstract

The microstructure evolution of ultrasonic additive manufactured (UAM) zirconium under room temperature uniaxial tensile straining is reported. Miniature dog-bone tensile specimens of two orientations were cut from a UAM zirconium bar for in situ synchrotron tensile tests. Wide-angle X-ray scattering (WAXS) scanning at the Advanced Photon Source (APS) at Argonne National Laboratory was used to unveil the changes in microstructure of the entire gauge regions throughout the straining. A series of WAXS data analysis methods were utilized to quantify both elastic and plastic deformation mechanisms within the strained specimens. Stress concentrations were identified during early stage of plastic deformation, which become candidate necking positions and eventually lead to failure. Fracture surface analysis implied that these stress concentration locations may be correlated to the fabrication defects, providing insightful guidance for future improvement of the UAM zirconium process.

Table of Contents

Abstract	v
Table of Contents.....	vi
1 Introduction.....	7
2 Experiments.....	8
2.1 Sample Summary.....	8
2.2 In Situ Synchrotron Tensile Test.....	9
2.3 Data Analysis.....	11
3 Results.....	14
3.1 Macroscopic Tensile Behavior.....	14
3.2 General WAXS Data.....	15
3.3 Microstructure Analysis on Elasticity.....	18
3.4 Microstructure Analysis on Plasticity.....	20
3.5 Fracture Surface Morphology.....	25
4 Discussion	26
5 Conclusions.....	28
Acknowledgements	28
References.....	29

1 Introduction

Additive manufacturing (AM) is a game-changing technology that make it feasible and economical to process components with complex geometry, which provide theoretically optimal performance according to advanced computer simulations [1,2]. The majority of metal AM methods, such as selective laser melting (SLM), require high processing temperature that may induce unfavorable thermal effects into the metals, leading to performance degradation compared to conventional manufacturing methods. On the other hand, combining advantages of both additive and subtractive processes, ultrasonic additive manufacturing (UAM) is a solid-state metal manufacturing approach that generates near net-shape components without involving unfavored thermal effects of laser-based metal AM technologies [3]. Therefore, UAM is particularly advantageous for producing parts made of temperature-sensitive materials.

Zirconium alloy is widely used in nuclear industry because it is a class of metallic structural materials that can provide acceptable material properties under high-temperature conditions in nuclear reactors without inducing extraneous neutronics penalty [4,5]. However, the preferred mechanical properties of Zr alloy originate from delicate heat treatment processes to ensure appropriate second phase precipitation, which may be compromised by AM process involving high temperatures [6]. Additionally, as a well-known getter material, Zr is reactive especially at high temperatures [7]. As a result, using laser-based AM processes for Zr may require inert atmospheres. Therefore, UAM, a low-temperature solid-state process, is an ideal approach to promote applications of Zr alloys in advanced nuclear reactor concepts.

Additionally, UAM has its unique capability of embedding sensors and detectors within the components, which could be powerful tools to enable validation of high-fidelity multiphysics reactor models and advanced reactor control technologies [8]. In order to apply UAM Zr alloy components in future advanced reactor designs, UAM Zr's microstructure characteristics and their corresponding effects on mechanical properties must be well-understood. In particular, UAM Zr consists of solid-state bonded Zr foils, which differ significantly from Zr alloy component fabricated using conventional metal processing approaches. Therefore, the microstructure evolution and failure mechanisms of UAM Zr are expected to be dissimilar to conventional Zr alloys and need to be investigated to guide UAM process optimization as well as inform future qualification and licensing efforts.

Featuring high intensity and high energy, the X-ray at the Advanced Photon Source (APS) at Argonne National Laboratory is capable of characterizing the macroscopic UAM Zr specimens [9]. Using in situ synchrotron wide-angle X-ray scattering (WAXS), a series of microstructure evolution information related to both elastic and plastic deformation can be unveiled by interpreting the diffraction information collected from a strain specimen. In the past decade, in situ synchrotron tensile tests have been successfully applied to a series of different advanced structural materials [10–17].

In this study, in-situ synchrotron tensile tests were performed on UAM Zr specimens of two different orientations at room temperature to examine the microstructure evolution of UAM Zr under deformation and identify its failure mechanism to inform future improvement efforts. The UAM-fabricated Zr samples were provided by Oak Ridge National Laboratory. The samples used in this work represent very early, preliminary samples from this process.

2 Experiments

2.1 Sample Summary

The UAM Zr specimens were obtained from Oak Ridge National Laboratory. According to previous tensile studies on UAM specimens, the normal direction (ND) specimens have poor tensile properties due to the premature debonding on foil interfaces [18]. Thus, the application of UAM components need to prevent tensile stress that is normal to the bonding interfaces. In this study, the transverse and rolling directions (TD/RD) specimens were the focus. In order to test the UAM Zr using the existing experimental setup, miniature dog-bone tensile specimens were prepared using electrical discharge machining (EDM) from the UAM Zr sample using both TD and RD orientations. EDM was selected because it does not induce excessive deformation into the specimens to affect original microstructure from UAM process. The dimensions of the miniature specimen can be found in Figure 1. For each tensile specimen, there is an approximately 5 mm (L) $\times$ 1.2 mm (W) $\times$ 0.5 mm (T) gauge region. The UAM Zr specimens grain size was characterized, and it was shown to contain grains that have a bimodal size distribution. Within each layer, the grain size is micron level (rolled Zr sheets). Between layers (solid state welding part), the grain size can be submicron. Synchrotron WAXS analysis can only handle up to $\sim$ 250 nm grain size. However, as both micron and submicron grains coexist, so it cannot distinguish it.

2.2 In Situ Synchrotron Tensile Test

The in-situ synchrotron WAXS tensile investigations were performed at Sector 1-ID-E of the Advanced Photon Source (APS) at Argonne National Laboratory (ANL). A 80.725 keV ($\lambda = 0.154 \text{ \AA}$) monochromatic synchrotron beam with a $100 \text{ }\mu\text{m} \times 100 \text{ }\mu\text{m}$ square beam shape was used to illuminate the gauge regions of the specimens to generate scattering signals. These scattering signals were collected by the Hydra WAXS detector array that consists of four GE-RT41 high-resolution ($2048 \text{ pixels} \times 2048 \text{ pixels}$) detectors. During each tensile test, the stage holding the tensile device reciprocated in the vertical direction so that 56 axial positions alongside the gauge region with 0.1 mm spatial intervals were scanned. It took approximately 100 seconds to take a set of WAXS exposures for the 56 axial positions. From the top of the scanned position to the bottom one, these axial positions were indexed from 1 to 56 sequentially. In order to ensure that each set of the WAXS exposures were collected under a similar tensile condition, a slow strain rate of $6 \times 10^{-6} \text{ s}^{-1}$ was adopted so that the microstructure evolution within one scan is insignificant. Two UAM Zr specimens of different orientations (i.e., TD and RD, see Figure 1a) were investigated using the aforementioned approaches. An overview of the experiment setup can be found in Figure 1b.

Figure 1. Tensile specimens investigated in this study and synchrotron experiment setup at APS: (a) a dog-bone tensile specimen and schematics showing the difference between TD and RD, and (b) overview of the experiment setup.

After the tensile test, the fracture surfaces of the two specimens were examined using an FEI Strata 400 DualBeam device equipped with a 30 kV field emission gun (FEG) scanning electron microscope (SEM). The fracture surface morphology was investigated to confirm the failure mechanisms identified using the synchrotron analysis.

2.3 Data Analysis

For each of the four GE-RT41 detectors, approximately six 10° azimuthal intervals of the Debye-Scherrer diffraction rings can be collected from the WAXS signals. These 24 intervals of WAXS information was azimuthally integrated to one-dimensional diffraction data (i.e., intensity versus diffraction angle), which were further fitted to pseudo-Voigt peak functions to obtain lattice spacings and peak breadths throughout the tensile process. The lattice spacings of 24 different azimuth angles were fitted to the following equation to deduce lattice strains:

$$d(\varphi) = \frac{d_y - d_x}{2} \cos(2\varphi) + \frac{d_y + d_x}{2} \quad (1)$$

where, φ is the azimuthal angle, d_y is the lattice spacing in the tensile or vertical direction ($\varphi = 0^\circ$ or 180°), and d_x is the lattice spacing in the horizontal direction ($\varphi = 90^\circ$ or 270°). On the other hand, the breadth of each reflection was deduced by calculating the average breach of the 24 intervals weighted by integrated intensity. At room temperature, Zr metal is dominated by α -phase that has a hexagonal closest packing (hcp) structure. Fourteen reflections of α Zr phase (i.e., $\{10\bar{1}0\}$, $\{0002\}$, $\{10\bar{1}1\}$, $\{10\bar{1}2\}$, $\{11\bar{2}0\}$, $\{10\bar{1}3\}$, $\{20\bar{2}0\}$, $\{11\bar{2}2\}$, $\{20\bar{2}1\}$, $\{0004\}$, $\{20\bar{2}2\}$, $\{10\bar{1}4\}$, $\{20\bar{2}3\}$, and $\{1\bar{1}20\}$) were involved in the WAXS data analysis. The lattice stains of these reflection were calculated using the following expressions:

$$\varepsilon_{yy}^{hkl} = \frac{d_y^{hkl} - d_0^{hkl}}{d_0^{hkl}} \quad (2)$$

$$\varepsilon_{xx}^{hkl} = \frac{d_x^{hkl} - d_0^{hkl}}{d_0^{hkl}} \quad (3)$$

where, ε_{xx}^{hkl} and ε_{yy}^{hkl} are the lattice strains of the $\{hkl\}$ reflection in horizontal and vertical directions, respectively; d_0^{hkl} is the stress-free lattice spacing of the $\{hkl\}$ reflection. The bulk lattice strain was then deduced by calculating the weighted average of the lattice strains of the 14 reflections using Daymond et al.'s approach [19].

In addition to the lattice strain evolution, which corresponds to the elastic deformation of the specimens, WAXS signals also contain information that correlates to the plastic deformation in the form of peak broadening. A series of peak broadening interpretation approaches were used in this study to quantify the plasticity based on WAXS peaking broadening, including the Williamson-Hall method (WH) [20], the modified Williamson-Hall method (mHW) [21], and extended Convolutional Multiple Whole Profile (eCMWP) [22]. The WH method assumes that the broadening of the diffraction peaks is contributed by two microstructure features: domain (grain) size and internal strain, which has the following form:

$$\beta \cos \theta = \frac{K\lambda}{D} + C\varepsilon \sin \theta \quad (4)$$

where, β is the breadth of the diffraction peak, θ is the diffraction angle, λ is the wavelength of the synchrotron photons, D is the average grain size, ε is the internal strain due to plastic deformation, K and C are the two adjustable constants that are usually 0.9 and 4.0, respectively. The breadth data of the 14 reflections were used to fit Equation (4) using linear regression. If the internal strain is assumed to be mainly contributed by dislocations, the mWH method can be used to quantify the dislocation density using the following expression:

$$\frac{\beta \cos \theta}{\lambda} = \frac{K}{D} + \left(\frac{\pi A^2}{2}\right)^{\frac{1}{2}} \rho^{\frac{1}{2}} (b^2 C)^{\frac{1}{2}} \left(\frac{2 \sin \theta}{\lambda}\right) \quad (5)$$

where, ρ is the dislocation density, b is the Burgers vector of dislocations, C is the contrast factor [23], A is an adjustable coefficient that was selected to be 1 for dislocation density around 10^{14} m^{-2} level. In hpc structure materials such as Zr, multiple slip systems with three different Burgers vectors (i.e., $\langle a \rangle$, $\langle c \rangle$, and $\langle a+c \rangle$) coexist. Therefore, the complex contrast-Burgers factor, $\overline{b^2 C}$ needs to be averaged over all possible systems. In this study, the approach used in Ungar et al.'s work [24] was used to develop average complex contrast-Burgers factor. The fractions of $\langle a \rangle$, $\langle c \rangle$ and $\langle a+c \rangle$ type dislocations were defined as h_a , h_c and h_{a+c} , which can be determined by maximize the fitting confidence parameter R^2 during linear regression of Equation (5). Moreover, the eCMWP was used to unveil more specific microstructure information, including both dislocation density and twinning probability using the approach reported by Balogh et al. [22] with appropriate modification to adapt Zr material. As eCMWP is an expensive computational process, only several representative positions and times were examined. For the WH and

mWH approaches, all 56 axial positions of the two strained samples throughout the straining process were analyzed.

At APS Sector 1-ID-E, ion chambers were used to measure the intensities of both incident (I_0) and transmitted (I_t) synchrotron beams. As synchrotron intensity attenuation in a Zr specimen follows exponential law as follows:

$$\frac{I_t}{I_0} = \exp(-\mu t) \quad (6)$$

where μ is the attenuation coefficient, and t is the thickness of the specimen. Therefore, using the maximum μt measured throughout the investigation of one specimen as the reference, normalized specimen thickness was calculated using the following formula so that the thinning effect can be captured:

$$t_N = -\frac{\ln(I_t / I_0)}{(\mu t)_{\max}} \quad (7)$$

3 Results

3.1 Macroscopic Tensile Behavior

The strain-stress curves of the two UAM Zr specimens with different orientations (TD and RD) are illustrated in Figure 2. It is prominent that the TD specimen has a slightly higher yield strength (YS: 380 MPa versus 350 MPa) and ultimate tensile strength (UTS: 481 MPa versus 463 MPa) compared to the RD specimen. Meanwhile, the elongation of the RD specimen is larger than that of the TD specimen (18.2% versus 16.1%). The difference in tensile properties between the TD and RD samples may originate from the different volume fraction of the interface-associated zone within the gauge region, which is discussed in detail later in this report.

Figure 2. Strain-stress curves of the RD and TD UAM Zr specimens measured in the in-situ experiment.

3.2 General WAXS Data

As the dog-bone specimens were shoulder-mounted onto stainless steel grips during the tensile test, only the gauge region was not blocked by the grips during the scanning. As shown in the left half of Figure 3, only scans 13 through 54 were not affected by the grip in the beginning of the tensile test, which can be defined as the field of view (FOV). Beyond this gauge region or FOV, the WAXS signals become weak and overlapped with diffraction information of stainless steels, affecting the quality of related data analysis results. The azimuthally integrated WAXS data of scan 1 and scan 30 of the RD specimens are illustrated in as representatives of WAXS signals out of and within the FOV, respectively. It is clear that the WAXS signals are overwhelmingly dominated by the diffraction from αZr within the FOV, while the stainless steel diffractions are the major signals in the WAXS signals out of the FOV.

Figure 3. A schematic drawing of the field of view (FOV) expansion during the tensile test.

Figure 4. The azimuthally integrated WAXS data of scan 1 and 30 of the RD specimen before the tensile test.

However, as the tensile test continues, the nature of the straining elongates the distance between the two grips. As a result, the FOV expands during the tensile straining. Near the end of the tensile test, 54 scanned positions (1 through 54) are within the FOV. In the experiment setup used in this study, the tensile device strains the specimen using an overhead pulling grip, while the bottom grip is fixed. Hence, as indicated in the right half of Figure 3, the bottom scans (high-index) correspond to the same physical positions of the specimen throughout the tensile test, while the top scans (low-index) correspond to different physical positions of the specimen as the gauge region elongates.

This FOV expansion can also be visualized by plotting the time evolution of normalized specimen thickness, as shown in Figure 5. The bottom grip was always captured by scan 55 throughout the tensile straining. On the other hand, in the beginning of the tensile straining, the top grip corresponded to scan 13. Immediately before specimen failure, the top grip moved to the domain of scan 1 or 2, indicating the expansion of the FOV. In Figure 5, it is clear that the gauge regions of both RD and TD specimens had

uniform thickness before straining. As straining continued, the specimens became thinner due Poisson's effect. In both RD and RD specimens investigated in this study, at least two axial positions were distinguished in each specimen to experience preferential thinning starting from the early stage of plastic deformation, which are marked in Figure 5 as red and blue dashed lines. The thickness evolution of these positions are compared with a reference position in each specimen (marked in Figure 5 as purple dashed lines) in Figure 6. One of these preferential thinning positions eventually evolved into the necking and failure position, as marked by the red arrow in each subfigure of Figure 5. Meanwhile, the other preferential thinning position also experienced minor necking effects as marked by the green arrow in the same subfigure, which will be referred to as the competing necking position. Hence, it is worth summarizing here that there are three representative positions in each specimen in this study: necking/failure position (red), competing necking position (green), and necking-free reference position (purple). In Figure 6, the difference in thickness between the failure and competing necking positions are only prominent after the true strain exceeds 0.10, implying that the competition for final failure position concludes when true strain is approximately 0.1.

Figure 5. Evolution of the normalized specimen thickness of the two strained specimens showing the formation of thin areas in the specimens with uniform original thickness. The failure positions are marked by red arrows; the non-failure thin areas are marked by green arrows; and the reference areas are marked by purple arrows.

Figure 6. Evolution of normalized specimen thickness at three representative axial positions versus true tensile strain: (a) RD specimen; (b) TD specimen.

3.3 Microstructure Analysis on Elasticity

The elastic deformation in the investigated specimens can be measured by the lattice strain information based on the diffraction peaking shifting in WAXS data. The average lattice over 14 α Zr reflections strain of the RD and TD specimens is shown in Figure 7. Elastic stress concentration was found at multiple axial positions in both sample after the specimen yielded. These stress concentration positions match those preferential thinning positions as shown in Figure 5, indicating a strong correlation between elastic stress concentration and necking/failure.

Figure 7. Evolution of average lattice strain of the two strained specimens showing the formation of elastic stress concentration areas. The failure positions are marked by red arrows; the non-failure thin areas are marked by green arrows; and the reference areas are marked by purple arrows.

The lattice strain of the three representative axial positions of each specimens are plotted separated in Figure 8. In both RD and TD specimens, starting from yielding, both failure and competing necking positions experience higher elastic deformation compared to the reference position. As necking initiated around 0.1 true strain, lattice strain of the failure position, which won the necking competition, continued to increase until failure occurred. On the other hand, lattice strain for both reference and competing necking positions started dropping as necking initiated. Throughout the tensile test, the reference position in both RD and TD specimens had a maximum lattice strain of approximately 0.0040, while the competing necking position reached approximately 0.0045 in lattice strain. The necking/failure positions of the two specimens, on the other hand, have lattice strain beyond 0.0060.

Figure 8. Evolution of averaged lattice strain at three representative axial positions versus true tensile strain: (a) RD specimen; (b) TD specimen.

3.4 Microstructure Analysis on Plasticity

As the grain size of the UAM Zr specimens investigated in this study is beyond the applicable range of the synchrotron X-ray setup, only the slope parts of the WH and mWH methods were interpreted in this paper. Using the WH method, the internal strain evolution, which is a measure of plastic deformation, is deduced for the RD and TD specimens throughout the tensile strain. In Figure 9, the fitting confidence (R^2) of the linear regression of Equation (4) are provided with the fitting results (i.e., internal strain). Except for those positions blocked by the top grip, which are ignored in this study, the WH fitting of the in situ WAXS data produced internal strain data with acceptable reliability. Similar to the findings of lattice strain, the internal strain of the RD and TD specimens was also found to have several localized axial positions, which corresponds the preferential thinning areas in Figure 5. That is, those axial positions showing preferential thinning during tensile strain simultaneously suffer localized lattice strain (elasticity) and internal strain (plasticity).

Figure 9. Evolution of internal strain of the two strained specimens deduced by the WH method along with corresponding fitting confidence (R^2), showing the formation of plastic deformation concentration areas. The failure positions are marked by red arrows; the non-failure thin areas are marked by green arrows; and the reference areas are marked by purple arrows.

Figure 10 Evolution of internal strain at three representative axial positions versus true tensile strain: (a) RD specimen; (b) TD specimen.

Similar to the approaches used previously, for each specimen, the internal strain of three representative axial positions are plotted separately in Figure 10 for detailed comparison. In both RD and TD specimens, those positions that are not involved with preferential thinning have an internal strain up to approximately 0.0011, while the competing necking positions have an ultimate internal strain of approximately 0.0013. On the other hand, once necking initiated, the localized internal strain near the failure point could approach 0.0020.

Assuming that the internal strain described above is mainly contributed by dislocations, the internal strain evolution shown in Figure 9 can be converted into dislocation density evolution using the mWH method, as indicated in Figure 11. Here, dislocations belonging to all the eleven slip systems of hcp structure materials are counted. In the mWH method, contrast factors are reflection dependent and slip system dependent. Therefore, as shown in Figure 11(a)(b), the fitting confidence of the mWH fitting is generally higher than that of the WH fitting in Figure 9(a)(b). Again, for each specimen, the total dislocation density evolution is plotted in detail for three representative axial positions in Figure 12. Prior to the tensile test, the dislocation density is $\sim 7.0 \times 10^{13} \text{ m}^{-2}$ and $\sim 8.0 \times 10^{13} \text{ m}^{-2}$ for the tested RD and TD specimens, respectively. The dislocation density in both specimens keeps constant before yielding, which is consistent with the definition. After yielding, the slip systems are activated so that dislocation propagation starts. At the reference axial positions in both samples, the dislocation density increases up to $\sim 5.5 \times 10^{14} \text{ m}^{-2}$. Meanwhile, at these competing necking positions, the ultimate total dislocation density is higher than that of the reference positions by $\sim 30\%$. Near the failure positions, the total dislocation density was found to be as high as $1.2 \times 10^{16} \text{ m}^{-2}$ in both specimens, which are the maximum values throughout the specimens.

Using the mWH approach adopted in this study, the dislocations with different Burgers vectors (i.e., $\langle a \rangle$, $\langle c \rangle$, and $\langle a+c \rangle$ for hcp system) can be distinguished by maximizing the fitting confidence. Hence, the density values of different types of dislocations of the representative axial positions are plotted separately in Figure 13 for both RD and TD specimens.

Figure 12. Evolution of total dislocation density at three representative axial positions versus true tensile strain: (a) RD specimen; (b) TD specimen.

Figure 13. Density evolution of different types of dislocations at three representative axial positions versus true tensile strain: (a) <a>-type dislocations in the RD specimen; (b) <a>-type dislocations in the TD specimen; (c) <c>-type dislocations in the RD specimen; (d) <c>-type dislocations in the TD specimen; (e) <a+c>-type dislocation sin the RD specimen; (f) <a+c>-type dislocations in the TD specimen.

The majority of the dislocations in both specimens are $\langle a \rangle$ -type. Before the tensile test, approximately 75% to 80% of the preexistent dislocations are $\langle a \rangle$ -type. As dislocations propagate during straining, the fraction of the $\langle a \rangle$ -type dislocations is stabilized at $\sim 70\%$. Therefore, the evolution of the $\langle a \rangle$ -type dislocations (Figure 13(a)(b)) are comparable to the total dislocation density (Figure 12). However, at the failure position, density of $\langle a \rangle$ -type dislocations becomes flattened during necking as it reaches $\sim 7.5 \times 10^{14} \text{ m}^{-2}$. As a result, the fraction of the $\langle a \rangle$ -type dislocations decrease to $\sim 60\%$ near the failure position before fracture. On the other hand, the evolution of the $\langle c \rangle$ -type dislocations as shown in Figure 13(c)(d) differ from that of the $\langle a \rangle$ -type dislocations. First of all, after yielding occurs, the $\langle c \rangle$ -type dislocations propagate after $\langle a \rangle$ -type dislocations. Also, the $\langle c \rangle$ -type dislocation density experiences a sharp drop at the late stage of necking near the failure position. Last, as indicated in Figure 13(e)(f), the $\langle a+c \rangle$ dislocation density increases slightly in the beginning of plastic deformation and then drops under the detection limit of the mWH method. However, at the failure positions, density of the $\langle a+c \rangle$ dislocations promptly increases at the late stage of necking.

Additionally, using the eCMWP code [25–27], the specimens were almost free of twinning before the tensile test. As the specimens are strained to failure, twinning probability in both specimens was found to be $\sim 0.04\%$ near the failure positions and $\sim 0.09\%$ in the reference positions. Meanwhile, eCMWP predicted dislocation density comparable to the mWH method. Therefore, dislocation propagation is the major plastic deformation mechanism in the UAM Zr specimens investigated in this study.

3.5 Fracture Surface Morphology

The fracture surfaces of the strained dog-bone specimens were investigated using SEM. As shown in Figure 14, the fracture surface morphology is dominated by dimples, implying that the specimen failure was caused by microvoid formation and growth under tensile stress. In addition, large parallel cracks can also be identified in the SEM images of the fracture surface, as marked by blue arrows in Figure 14. The spacing between these parallel cracks are close to the foil thickness used for UAM process. Therefore, these cracks should be the interface cracking. Therefore, failure of UAM bonding interfaces plays an important role in the UAM Zr failure.

Figure 14 SEM fracture surface of the RD UAM Zr specimen.

4 Discussion

Conventional polycrystalline Zr has YS of ~310 MPa and UTS of ~370 MPa, which are lower than the UAM Zr specimens of both orientations tested in this study [28]. Meanwhile, the strengthening effect of the UAM Zr comes with a cost in reduced elongation. The ~16% and ~18% elongation of the UAM specimens are prominently lower than that of the conventional Zr (~21%) [28]. The difference in mechanical properties between the UAM Zr and conventional polycrystalline Zr are speculated to originate from the bonding interfaces formed during UAM process. As a solid-state AM process, UAM joins metal foils through recrystallization across the interface, the microstructure and mechanical properties near the interface (i.e., interface regions) are different from those of the bulk foil regions. In addition, the preexistent surface oxidation of the foil may also induce interface oxide impurity in UAM specimens [29]. Therefore, the interface region is expected to have higher YS and UTS but lower ductility due to both reduced grain size [30] and oxide inclusions. This may help explain the dissimilar mechanical properties of UAM Zr specimens compared to conventional polycrystalline Zr. Also, the major difference between the RD and TD dog-bone specimens is the number of foil layers and consequently the volume fraction of interface regions. This may eventually lead to the different macroscopic mechanical properties as shown in Figure 2.

On the other hand, a major finding of this in situ WAXS tensile investigation is the multiple preferential thinning positions, which eventually lead to the failure of the sample. These thinner regions cannot be distinguished before straining and are only prominent after yielding. They also correlate to stress concentration and localized plasticity as unveiled by WAXS lattice strain and WH/mWH analyses. During plastic deformation, these preferential thinning positions compete with each other until prominent necking phenomenon occurred at one of them. It is speculated that those candidate necking locations are correlated to high local density of manufacturing defects, which are mainly interface voids arising from the UAM process. Interface voids are common manufacture defects of the UAM method due to local ineffective joining [31]. These interface voids are usually elongated so that the effective thickness (which can be detected by synchrotron transmission) is barely affected. However, these interface defects are equivalent to local debonding between foils, causing stress concentration under tensile straining and eventually preferential dislocation propagation and microvoid propagation. Meanwhile, these interface voids also initiate the formation of interface cracks as local stress is high enough, which were confirmed by SEM images of the fracture surface. Hence, based on the WAXS analysis, interface voids were found to be the source of the main failure mechanism of these UAM Zr specimens. Therefore, reducing interface voids will be an efficient approach to improve the mechanical properties of UAM Zr. Post-fabrication treatment methods, such as hot isostatic pressing (HIP), were found to help eliminating these interface voids [31]. However, as Zr is a heat-sensitive structural material, the utilization of post-fabrication treatment could be restricted. Alternative mitigation approaches may include pre-processing of the foil surface and optimization of the UAM parameters.

Last but not least, it is worth mentioning that this investigation was performed at room temperature. At elevated temperatures, the anticipated service window for UAM Zr, the deformation mechanisms are expected to change. Therefore, high temperature in situ synchrotron WAXS tensile investigations are being planned for these UAM Zr specimens so that a more systematic understanding of the performance of this innovative material can be assessed.

5 Conclusions

In this study, preliminary ORNL-provided UAM Zr specimens of both RD and TD orientations were investigated at the APS Facility at Argonne National Laboratory using in situ synchrotron WAXS tensile test at room temperature. A series of WAXS analysis methods were adopted to retrieve a variety of quantitative microstructure evolution information, including thickness, elasticity and plasticity, throughout the gauge regions of the tensile specimens. The UAM Zr was found to provide higher mechanical strength compared to conventional polycrystalline Zr with slight compromise in ductility. Based on WAXS analysis, localized stress concentration and preferential plasticity and thinning were identified to be the major failure mechanism of this set of UAM Zr specimens, which originates from the interface voids formed during UAM process. This finding provides insightful guidance for future improvement of UAM Zr technology.

Acknowledgements

This work was sponsored by the U.S. Department of Energy, Office of Nonproliferation Research and Development in the U.S. National Nuclear Security Administration Office of Defense Nuclear Nonproliferation under Contract DE-AC02-06CH11357. The samples used in this study were provided by Oak Ridge National Laboratory.

References

- [1] W.E. Frazier, Metal additive manufacturing: a review, *J. Mater. Eng. Perform.* 23 (2014) 1917–1928.
- [2] T.D. Ngo, A. Kashani, G. Imbalzano, K.T.Q. Nguyen, D. Hui, Additive manufacturing (3D printing): A review of materials, methods, applications and challenges, *Compos. Part B Eng.* 143 (2018) 172–196.
- [3] M. Norfolk, H. Johnson, Solid-state additive manufacturing for heat exchangers, *JOM.* 67 (2015) 655–659.
- [4] D.O. Northwood, The development and applications of zirconium alloys, *Mater. Des.* 6 (1985) 58–70.
- [5] R. Krishnan, M.K. Asundi, Zirconium alloys in nuclear technology, *Proc. Indian Acad. Sci. Sect. C Eng. Sci.* 4 (1981) 41–56.
- [6] A.R. Massih, T. Andersson, P. Witt, M. Dahlbäck, M. Limbäck, Effect of quenching rate on the β -to- α phase transformation structure in zirconium alloy, *J. Nucl. Mater.* 322 (2003) 138–151.
- [7] W. Espe, M. Knoll, M.P. Wilder, Getter materials, *Electronics.* 23 (1950) 80.
- [8] A. Hehr, M. Norfolk, J. Sheridan, M. Davis, W. Leser, P. Leser, J.A. Newman, Spatial strain sensing using embedded fiber optics, *JOM.* 71 (2019) 1528–1534.
- [9] J. Lin, X. Han, B.J. Heuser, J.D. Almer, Study of the mechanical behavior of the hydride blister/rim structure in Zircaloy-4 using in-situ synchrotron X-ray diffraction, *J. Nucl. Mater.* 471 (2016) 299–307.
- [10] X. Liu, K. Mo, Y. Miao, K.-C. Lan, G. Zhang, W.-Y. Chen, C. Tomchik, R. Seibert, J. Terry, J.F. Stubbins, Investigation of thermal aging effects on the tensile properties of Alloy 617 by in-situ synchrotron wide-angle X-ray scattering, *Mater. Sci. Eng. A.* 651 (2016). <https://doi.org/10.1016/j.msea.2015.10.098>.
- [11] Y. Miao, K. Mo, X. Liu, Z. Zhou, J. Aimer, J.F. Stubbins, Response of oxide particles to externally applied stress in austenitic ODS alloys, in: *Trans. Am. Nucl. Soc.*, 2014.
- [12] K. Mo, Y. Miao, Z. Zhou, D. Yun, X. Liu, J. Aimer, J.F. Stubbins, Nanoparticles loading behavior before and after matrix necking: An in-situ synchrotron radiation study in a 9Cr ODS alloy, in: *Trans. Am. Nucl. Soc.*, 2014.

- [13] J.-L. Lin, K. Mo, D. Yun, Y. Miao, X. Liu, H. Zhao, D.T. Hoelzer, J.-S. Park, J. Almer, G. Zhang, Z. Zhou, J.F. Stubbins, A.M. Yacout, In situ synchrotron tensile investigations on 14YWT, MA957, and 9-Cr ODS alloys, *J. Nucl. Mater.* 471 (2016). <https://doi.org/10.1016/j.jnucmat.2015.10.049>.
- [14] Y. Miao, K. Mo, Z. Zhou, X. Liu, K.-C. Lan, G. Zhang, M.K. Miller, K.A. Powers, Z.-G. Mei, J.-S. Park, J. Almer, J.F. Stubbins, On the microstructure and strengthening mechanism in oxide dispersion-strengthened 316 steel: A coordinated electron microscopy, atom probe tomography and in situ synchrotron tensile investigation, *Mater. Sci. Eng. A.* 639 (2015). <https://doi.org/10.1016/j.msea.2015.05.064>.
- [15] Y. Gan, K. Mo, D. Yun, D.T. Hoelzer, Y. Miao, X. Liu, K.-C. Lan, J.-S. Park, J. Almer, T. Chen, H. Zhao, Temperature effect of elastic anisotropy and internal strain development in advanced nanostructured alloys: An in-situ synchrotron X-ray investigation, *Mater. Sci. Eng. A.* 692 (2017). <https://doi.org/10.1016/j.msea.2017.03.068>.
- [16] X. Zhang, M. Li, J.-S. Park, P. Kenesei, J. Almer, C. Xu, J.F. Stubbins, In situ high-energy X-ray diffraction study of tensile deformation of neutron-irradiated polycrystalline Fe-9% Cr alloy, *Acta Mater.* 126 (2017) 67–76.
- [17] C. Tomchik, J. Almer, O. Anderoglu, L. Balogh, D.W. Brown, B. Clausen, S.A. Maloy, T.A. Sisneros, J.F. Stubbins, High energy X-ray diffraction study of the relationship between the macroscopic mechanical properties and microstructure of irradiated HT-9 steel, *J. Nucl. Mater.* 475 (2016) 46–56.
- [18] N. Sridharan, M. Gussev, R. Seibert, C. Parish, M. Norfolk, K. Terrani, S.S. Babu, Rationalization of anisotropic mechanical properties of Al-6061 fabricated using ultrasonic additive manufacturing, *Acta Mater.* 117 (2016) 228–237.
- [19] M.R. Daymond, The determination of a continuum mechanics equivalent elastic strain from the analysis of multiple diffraction peaks, *J. Appl. Phys.* 96 (2004) 4263–4272.
- [20] G.K. Williamson, W.H. Hall, X-ray line broadening from fcc aluminium and wolfram, *Acta Metall.* 1 (1953) 22–31.
- [21] T. Ungár, Á. Révész, A. Borbély, Dislocations and grain size in electrodeposited nanocrystalline Ni determined by the modified Williamson--Hall and Warren--Averbach procedures, *J. Appl. Crystallogr.* 31 (1998) 554–558.
- [22] L. Balogh, G. Tichy, T. Ungár, Twinning on pyramidal planes in hexagonal close packed crystals

- determined along with other defects by X-ray line profile analysis, *J. Appl. Crystallogr.* 42 (2009) 580–591.
- [23] I.C. Dragomir, T. Ungár, Contrast factors of dislocations in the hexagonal crystal system, *J. Appl. Crystallogr.* 35 (2002) 556–564.
 - [24] T. Ungár, O. Castelnau, G. Ribárik, M. Drakopoulos, J.L. Béchade, T. Chauveau, A. Snigirev, I. Snigireva, C. Schroer, B. Bacroix, Grain to grain slip activity in plastically deformed Zr determined by X-ray micro-diffraction line profile analysis, *Acta Mater.* 55 (2007) 1117–1127.
 - [25] G. Ribárik, T. Ungár, J. Gubicza, MWP-fit: a program for multiple whole-profile fitting of diffraction peak profiles by ab initio theoretical functions, *J. Appl. Crystallogr.* 34 (2001) 669–676.
 - [26] G. Ribárik, J. Gubicza, T. Ungár, Correlation between strength and microstructure of ball-milled Al-Mg alloys determined by X-ray diffraction, *Mater. Sci. Eng. A.* 387 (2004) 343–347.
 - [27] L. Balogh, G. Ribárik, T. Ungár, Stacking faults and twin boundaries in fcc crystals determined by x-ray diffraction profile analysis, *J. Appl. Phys.* 100 (2006) 23512.
 - [28] M. Rafique, N. Afzal, R. Ahmad, S. Ahmad, I.M. Ghauri, Mechanical behavior of low-dose neutron-irradiated polycrystalline zirconium, *Radiat. Eff. Defects Solids.* 167 (2012) 289–297.
 - [29] R.R. Dehoff, S.S. Babu, Characterization of interfacial microstructures in 3003 aluminum alloy blocks fabricated by ultrasonic additive manufacturing, *Acta Mater.* 58 (2010) 4305–4315.
 - [30] N. Hansen, Hall–Petch relation and boundary strengthening, *Scr. Mater.* 51 (2004) 801–806.
 - [31] M.N. Gussev, N. Sridharan, Z. Thompson, K.A. Terrani, S.S. Babu, Influence of hot isostatic pressing on the performance of aluminum alloy fabricated by ultrasonic additive manufacturing, *Scr. Mater.* 145 (2018) 33–36.

Nuclear Science & Engineering Division

Argonne National Laboratory
9700 South Cass Avenue, Bldg. 208
Argonne, IL 60439

www.anl.gov

U.S. DEPARTMENT OF
ENERGY

Argonne National Laboratory is a U.S. Department of Energy
laboratory managed by UChicago Argonne, LLC