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Radiation Induced Segregation at Low Angle Grain Boundaries in Steels: NSUF 2017 Milestone Report

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OAK RIDGE NATIONAL LABORATORY
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Materials Science and Technology Division

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NSUF 2017 Milestone Report

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CONTENTS

LIST OF FIGURES

LIST OF TABLES

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ACRONYMS

ADF	Annular Dark Field
APT	Atom Probe Tomography
CAES	Center for Advanced Energy Studies
CINR	Consolidated Innovative Nuclear Research
DOE	Department of Energy
EBSD	Electron Backscattered Diffraction
EDS	X-ray Dispersive Spectroscopy
FIB	Focused Ion Beam
HAADF	High Angle Annular Dark Field
INL	Idaho National Laboratory
LAMDA	Low Activation Materials Development and Analysis
NE	Nuclear Energy
NSUF	Nuclear Scientific User Facility
ORNL	Oak Ridge National Laboratory
RIS	Radiation-Induced Segregation
SEM	Scanning Electron Microscope
STEM	Scanning Transmission Electron Microscope
TEM	Transmission Electron Microscope

EXECUTIVE SUMMARY

This document outlines the user supported post-irradiation characterization by Oak Ridge National Laboratory under the Nuclear Scientific User Facility Consolidated Innovative Nuclear Research project, “Correlative Atom Probe and Electron Microscopy Study of Radiation Induced Segregation at Low and High Angle Grain Boundaries in Steels.” Fiscal Year 2017 was the first of a two-year project combining atom probe tomography and analytical electron microscopy. Electron Backscatter Diffraction was performed on the specimens to locate high and low angle grain boundaries for specimen prepared using focused ion beam. Focused ion beam prepared lift outs were used for analytical electron microscopy of the grain boundaries using the FEI F200X Talos. This report briefly summarizes the specimen preparation and analytical electron microscopy results obtained at Oak Ridge National Laboratory’s Low Activation Materials Development and Analysis facility.

1. INTRODUCTION

In 2016, Oak Ridge National Laboratory (ORNL) principal investigators successfully proposed to develop a more robust understanding of the nature of defect sinks under irradiation by exploring grain boundaries in full 3D with spatial resolution less than one nanometer. The DOE Office of Nuclear Energy (NE) Nuclear Science User Facility (NSUF) provided user supported access to a combination of key facilities at ORNL and Idaho National Laboratory (INL) capable of providing scanning transmission electron microscopy (STEM) with ultra-high efficiency x-ray dispersive spectroscopy (EDS) using a 360° rotation holder and atom probe tomography (APT) on irradiated austenitic core shroud materials. By studying the nature of sink-matrix interfaces, the principle investigators seek to demonstrate new techniques to gain a wider fundamental understanding of nuclear materials under elevated temperature irradiation.

The Consolidated Innovative Nuclear Research (CINR) grant provided funded access to ORNL's Low Activation Materials Development and Analysis (LAMDA) facility and laboratory technicians to identify, prepare, and survey specimens for use in the LAMDA microcopy facility. It also funds the user support on instruments including the focused ion beam (FIB) and transmission electron microscope (TEM). Austenitic 304 core shroud and comparative high-purity E-alloy samples were selected by the principle investigators based on a review of the available material relevant to the study. This report documents the specimen preparation, electron backscatter diffraction, and characterization in successful completion of a PICS Milestone M3UF-17OR0207192 – "Complete Low Angle Grain Boundary APT and Electron Microscopy".

2. SPECIMEN IDENTIFICATION AND METALLOGRAPHY

The principle investigators identified 4 TEM disk samples of austenitic 304 core shroud material irradiated at 5.5, 10.2 and 47.5 dpa and comparative E-alloy low tramp element material irradiated to 10.2 dpa as shown in Table 1. All specimens had an irradiation temperature of 320°C. LAMDA facility technicians located the specimens and mounted them in the correct experimental geometry. The mounted specimens were moved to a dual-beam scanning electron microscopy (SEM) - focused ion beam (FIB) to complete microstructural characterization.

Table 1: Samples identified for characterization

Sample ID	Alloy	Specimen Geometry	Damage (dpa)
AS13	304C	TEM Disk	5.5
AS18	304C	TEM Disk	10.2
AS23	304C	TEM Disk	47.5
ES16	304C	TEM Disk	22.5

3. FIB-EBSD AND SPECIMEN PREPARATIONS

3.1 ELECTRON BACKSCATTER DIFFRACTION

Electron backscatter diffraction (EBSD) was performed in the ORNL LAMDA FEI Versa3D DualBeam SEM-FIB instrument (FEI, Hillsboro, OR, USA) for all 4 specimens. The EBSD system is a modified version of the Oxford NordlysMax high-efficiency platform for high speed data acquisition. The data was acquired with a 20 kV 4.0 nA beam at a tilt of 70^{o} with a step size of $\sim 0.5\ \mu m$. High angle and low angle grain boundaries were selected for FIB preparation on specimens AS13, AS18, AS23, and ES16. Once the desired grain boundaries were identified using EBSD, platinum straps were placed across the grain boundaries as markers for future lift-out positions and to protect the sub-surface portion of the TEM lift outs. Additional platinum straps were placed in-line with the high and low angle desired grain boundaries to identify atom probe lift out locations. Figure 1 shows EBSD imagery from specimens AS18, AS13, and AS23. A total of 4 platinum straps were laid down on each specimen, though not all survived the final FIB lift out procedures.

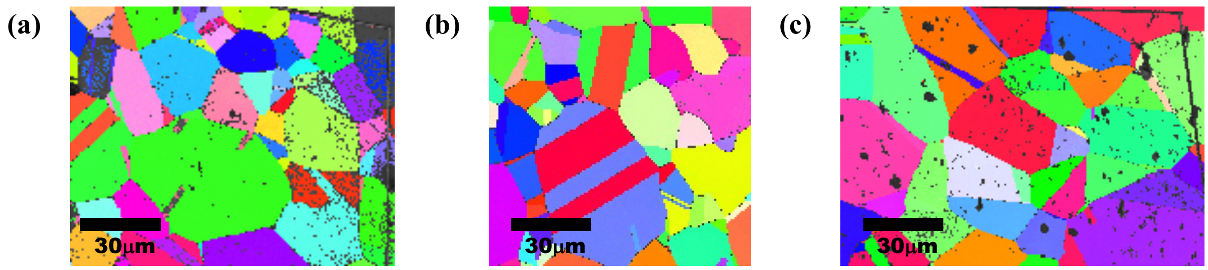

Figure 1: EBSD maps of CINR specimens (a) AS18, (b) AS13, (c) AS23.

3.2 PREPARATION OF TEM AND APT SAMPLES

After grain boundaries of interest were identified and marked, 8 TEM and 2 APT lift outs were created using a FEI Versa 3D DualBeam SEM-FIB and a FEI Quanta 3D 200i DualBeam SEM-FIB. TEM lift outs were taken across high and low angle grain boundaries and attached to molybdenum TEM grids and thinned to the desired thickness (<100 nm). Two APT lift outs were taken from specimen AS23 along the high angle and low angle grain boundaries. The lift outs were attached to pillars on silicone coupons made specifically for APT. An example of the high angle grain boundary TEM lift out of AS18 is shown in Figure 2 with the reference EBSD pattern.

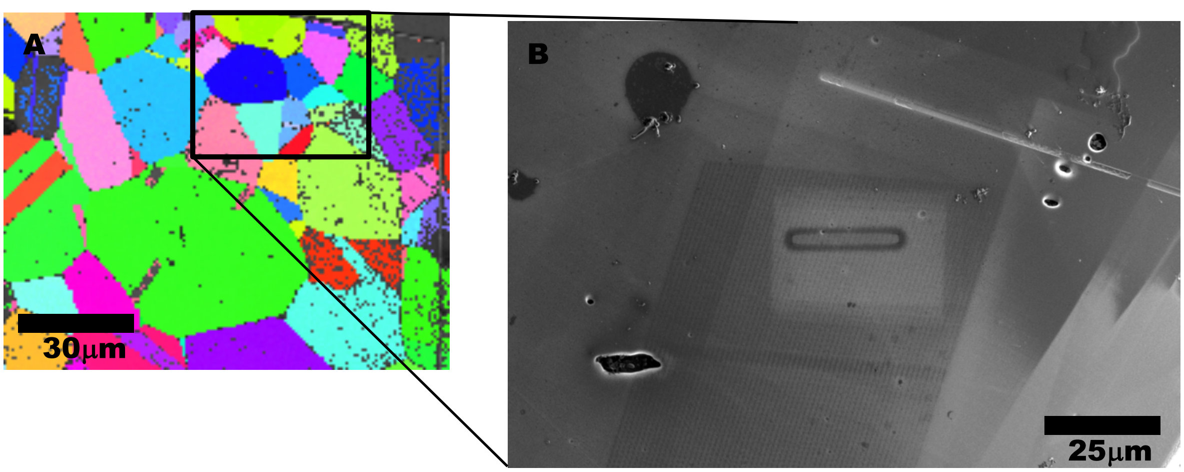

Figure 2: (a) AS18 EBSD map and (b) SEM image of high angle grain boundary TEM lift out.

4. TEM CHARACTERIZATION

FIB prepared specimens were analyzed using the 200 kV FEI Talos F200X STEM (FEI, Hillsboro, OR, USA). Table 2 summarizes the current status of the RIS analysis completed on the specimens prepared in Section 3.2. Several of the targeted lift-outs did not render viable samples and hence were not characterized during the first series of RIS analysis and quantification. For the acceptable samples, preliminary analysis has been completed using standard RIS analysis.

Table 2: Summary of specimens prepared and then analyzed using the FEI F200X Talos.

Grain Boundary Type	**Sample ID**		
	AS13	**AS18**	**AS23**
Low Angle	RIS analysis complete	No GB found in sample	Sample destroyed
High Angle	RIS analysis complete	Sample under prep.	No GB found in sample

Annular dark field (ADF)-High Angle Annular Dark Field (HAADF) image pairs and EDS spectrum images (1024×1024 pixels, 0.23 nm/pixel, 3600 s acquisition time) were completed on varying grain boundaries of interest. Quantification of the spectrum images was completed using the Bruker Quantax Esprit software using the Cliff-Lorimer quantification method. 1D-line profiles were extracted from the 2D quantitative maps by binning along the direction of the grain boundary.

The STEM-EDS analysis from the AS13 sample, Figure 3, shows the expected RIS analysis based on the previous findings by Field et al. [1]. Grain boundaries were enriched in Ni and Si while depleted in Fe and Cr. The spectrum images also show the formation of Ni-Si rich clusters observed within the grains of the specimens although a distinct denuded zone can be observed in the vicinity of both the high angle and low angle grain boundaries in the AS13 sample. The Ni-Si clusters are found to be randomly distributed throughout the matrix. The composition of the clusters cannot be conclusively determined due to matrix contributions within their EDS signals. This limitation necessitates the need for atom probe tomography to accurately quantify the composition. Given this, it is assumed the clusters are either γ′ (Ni_3Si) or G-phase ($M_6Ni_{16}Si_7$) based on insights from a previous study [2].

Line profiles across the grain boundaries in Figure 3 are shown in Figure 4. Line profiles show the expected 'w-profile' for the RIS response in this alloy based on previous work [1]. Interestingly, little difference is observed in the magnitude of the RIS response between the two different grain boundary types. Further work is needed to verify the EBSD results, such as Kikuchi pattern diffraction analysis, but the current set of results suggest the analyzed low angle grain boundary has sufficient misorientation (energy) to act as an efficient sink for irradiated defects within the characterized material. The present results suggest that future targeted lift-outs for atom probe tomography work needs to focus on very low angle (<5°) grain boundaries to easily detect RIS to the grain boundary dislocation cores.

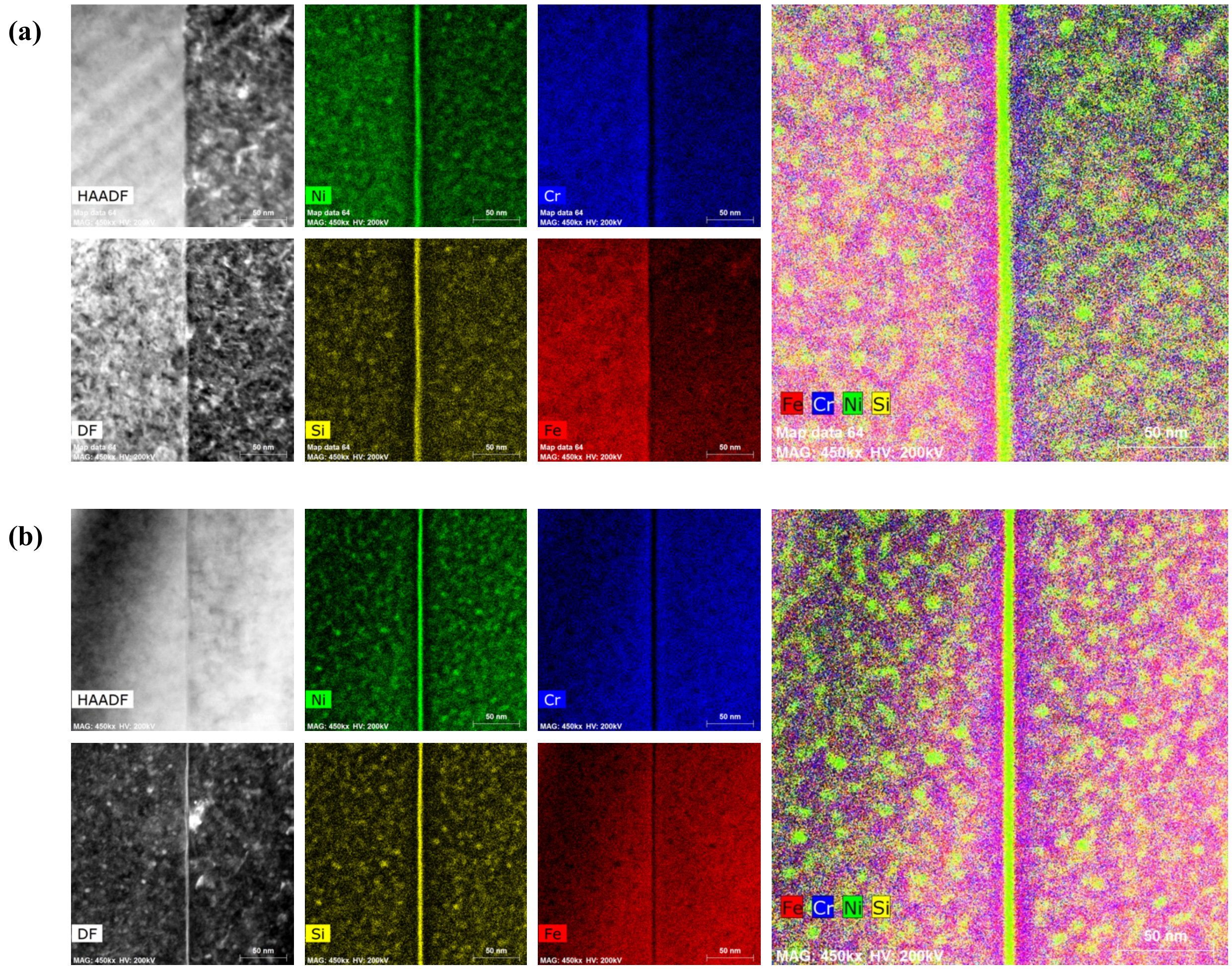

Figure 3: Annular Dark Field (ADF), High Angle Annular Dark Field (HAADF), elemental EDS maps, and elemental EDS overlay maps extracted from spectrum images showing the Ni-Si cluster formations and RIS in grain boundaries irradiated to the 5.5 dpa, 320°C condition; (a) low angle grain boundary, (b) random high angle grain boundary.

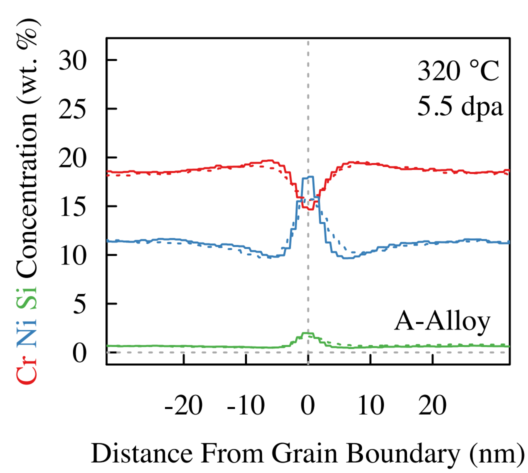

Figure 4: 1D RIS profiles for a random high angle grain boundary (solid line) and low angle grain boundary (dotted line) in the specimen irradiated to 5.5 dpa at 320°C.

5. CONCLUSION AND PATHWAY FORWARD

The successful completion of 2017 NSUF Milestone "Complete Low Angle Grain Boundary APT and Electron Microscopy" included user supported specimen identification, mounts, EBSD, FIB, and STEM-EDS. ORNL laboratory technicians and microscopy support staff prepared samples and provided 8 TEM lift outs and 2 APT needles to principle investigators to study grain boundaries using electron microscopy and atom probe tomography. The on-going user supported characterization in 2018 will focus on additional lift outs for 360° characterization and atom probe tomography.

6. REFERENCES

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