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Nuclear Science User Facilities: Instrument Scientist Program

October 2020

FY 2020 Progress Report

Dr. Jeffrey Giglio

Idaho National Laboratory

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ABSTRACT

The traditional single institution User Facility model prevalent in the DOE complex funds Instrument Scientists (IS) full-time to collaborate with and guide users, disseminate results through publication, build a broad and competent user base, continuously improve instrumentation, enhance data collection methods, and improve data analysis methods. This model focuses the Instrument Scientist on ensuring that visiting researchers obtain the best and most comprehensive data.

Instrument Scientists are also expected to establish personal research programs based on the use of the instrument(s) under their charge, which allows for further advances in instrumentation and science. This model ensures that the User Facility offers unique, world-leading instruments and data analysis to the user community, creates high demand among top researchers for the use of these capabilities, and as a result, positively impacts science and technology in the United States.

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1. INSTRUMENT SCIENTIST ACCOMPLISHMENTS

1.1 Introduction

The traditional single institution User Facility model prevalent in the Department of Energy (DOE) complex funds Instrument Scientists (IS) full-time to collaborate with and guide users, disseminate results through publication, build a broad and competent user base, continuously improve instrumentation, enhance data collection methods, and improve data analysis methods. This model focuses the Instrument Scientist on ensuring that visiting researchers obtain the best and most comprehensive data. Instrument Scientists are also expected to establish personal research programs based on the use of the instrument(s) under their charge, which allows for further advances in instrumentation and science. This model ensures that the User Facility offers unique, world-leading instruments and data analysis to the user community, creates high demand among top researchers for the use of these capabilities, and, as a result, positively impacts science and technology in the United States.

The Nuclear Science User Facilities (NSUF) has not historically operated using a traditional User Facility funding model. In the previous model, funding is provided to Instrument Scientists only to accomplish specific, narrowly defined tasks, associated with an awarded research project. Virtually no time is allocated for capability development. This model did not effectively address the underlying scientifically relevant issues associated with nuclear energy, including those related to the materials science of nuclear fuels and materials performance. It also results in instrument capabilities that remain largely static rather than increasing in the ability to produce impactful scientific results over time.

DOE-Nuclear Energy (NE) and the Idaho National Laboratory (INL) have worked to install significant scientific instrumentation at INL over the last decade. These investments have resulted in world-class nuclear energy research instruments. In order to best use these instruments, it was necessary to establish a cadre of Instrument Scientists with the breadth and depth of expertise necessary to ensure that these capabilities positively influence the future of nuclear energy through both incremental technical progress and significant technological breakthroughs. Starting in FY 2017, NSUF created a system to support instrument scientists to develop new capabilities on their instruments outside of their direct project work. Table 1 summarizes the scientists supported by NSUF in FY 2020 and their notable outcomes.

This year NSUF IS work was influenced by the curtailment of work at the INL due to the COVID-19 virus. The stoppage of work in the research facilities, the backlog of programmatic work and graded approach to restart activities has led several of the Instrument Scientists to have little contributions as compared to previous years.

Table 1. NSUF Instrument Scientist FY 2020. Scientists in bold denote New Hires in PIE Division (i.e., less than 1 year at INL).

Scientist	Area of Research	NSUF Support FTE	Notable Outcome for FY 2020
M. Bachhav	Atom Probe	0.2	3 publications
F. Di Lemma	EBSD sample preparation for air sensitive materials	0.2	2 publications submitted 1 publication in progress 1 white paper produced
L. He	Enhanced TEM characterization	0.2	3 publications
S. Middlemas	Thermal properties	0.2	1 publication 1 publication in progress
D. Murray	FIB Method Development	0.2	1 publication 2 publications submitted
X. Liu	Enhanced TEM characterization through software advances	0.2	2 publications 1 publication under review
C. Sun	Micro-mechanical testing	0.2	2 publications submitted
F. Cappia	Advanced Image Processing	0.2	2 publications in review
A. Craft	Neutron Imaging	0.2	1 publication Protoype for rad sample stage
N. Cordes	X-Ray, μ-CT	0.2	Irradiated Sample Handling in X-ray microscope
L. Squires/ L. Albuquerque	Np Production and Characterization	1	Produce Np metal “at will”
K. Wright	Electron Probe MicroAnalyzer	0.2	1 publication accepted 1 publication submitted
C. Tan	Positron Annihilation Spectroscopy	0.2	Setup and tested digital PAS system
N. Cinbiz	Digital Image Correlation	0.2	Setup and tested DIC stereoscope
Boopathy Kombaiah	PPMS	0.2	*(CoVID and Chiller Problem)*
Xiaofei Pu	XRD	0.2	Receive training on XRD

1.2 Dr. Mukesh Bachhav

Dr. Mukesh Bachhav has been developing advanced methods in atom probe tomography (APT) for the characterization of nuclear materials during FY 2020. Dr. Bachhav was funded at the level of 0.20 FTE during FY 2020. The focus of Dr. Bachhav’s work in FY 2020 were: Method development for improving the accuracy of cluster analysis by participating in a round robin carried out by Oxford University, University of Wisconsin and INL; improving the data interpretation and analysis methodology for thermal effects on the characterization of ceramic fuels; optimization of running conditions for quantification of minor solute element i.e., Zr-0.5Nb. Additional details are given below.

APT is a very powerful technique for studying clustering or the formation of very small precipitates that are often formed during irradiation and contribute to changes in material properties and corrosion behavior. Often, it is easy to see in the 3D atom maps if an element is clustered. However, to quantitatively determine the density of clusters and the number of atoms in each cluster is not trivial. The most used algorithm to obtain this data is the maximum separation method (MSM). A round robin exercise using the MSM algorithm revealed that different laboratories/users will not obtain the same results; the spread in data reported number of clusters, expressed as the standard deviation divided by the average, was typically 40%. This work demonstrates that a relatively large variation in results is likely when using the MSM algorithm without a standardized methodology, and highlights the need for a consensus standard to be developed. A detailed report of the cluster analysis of 3 different datasets involving clustering in Zr alloys was submitted to the MUZIC-3 program led by the Electric Power Research Institute (EPRI).

Table 2. Information about the three datasets.

No.	Material	Provider
1	Neutron-irradiated Zr-2.5Nb from CNL	Oxford
2	Proton-irradiated (1 dpa) Zr-0.5Nb	Wisconsin
3	Neutron-irradiated Zircaloy-2 from KKL	Chalmers

Figure 1. APT images of the three datasets analyzed in this study, 1 to 3 from left to right.

APT is not only useful for 3D distribution and chemical analysis of nuclear fuels and materials but also for quantitative isotopic analysis which provides insight into local fission densities (burnup) that may be affected by self-shielding or power peaking. Steep gradients in fission density that are present in some fuels lead to large differences in fuel microstructure that complicate interpretation of fuel performance. Isotopic analysis of ceramic fuels is often challenging for quantification of peaks due to the intrinsic thermal tails associated with analysis in laser mode. As shown in Figure 2 below, UO_2 has longer thermal tails which makes isotopic quantification very difficult in comparison to U-Mo metallic fuel which has a negligible ratio of thermal tails to peak intensity. Work is in progress to develop methods that can be used to quantify data that exhibit thermal tails to determine the relative distribution of U isotopes for evaluating local fission densities.

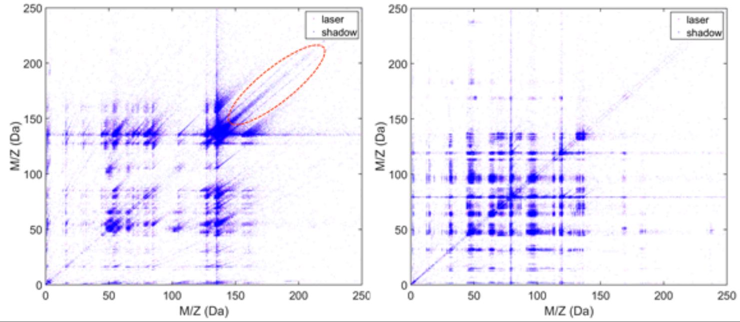


Figure 2. Correlative multihit analysis showing delayed evaporation (dotted lines) for UO2 fuel (left) in comparison to metallic U-Mo fuel (right).

Quantification of the Nb distribution in Zr alloys is extremely important for understanding irradiation effects but also for predicting corrosion behavior. However, choosing the right parameter to run these samples in APT is not straightforward. Systematic analysis was performed to evaluate the role of instrument operating parameters on achieving accurate chemical quantification of Nb. This study is still under investigation.

An important benefit of the IS program is the ability to collaborate with other atom probe groups around the world and build personal and collective expertise on the collection, analysis, and interpretation of atom probe data. These collaborations broadly advance the use of APT in understanding and solving nuclear fuel and material. Performance issues. A list of collaborators resulting in part from the IS funding is provided below:

1. Anderoglu group, University of New Mexico
2. Couet group, University of Wisconsin
3. Jadernas, Studsvik, Sweden
4. Wharry group, Purdue University
5. Getto, USNA
6. Odette, UCSB
7. Shridharan group, University of Wisconsin
8. Short group, MIT
9. Yang group, U. of Florida.

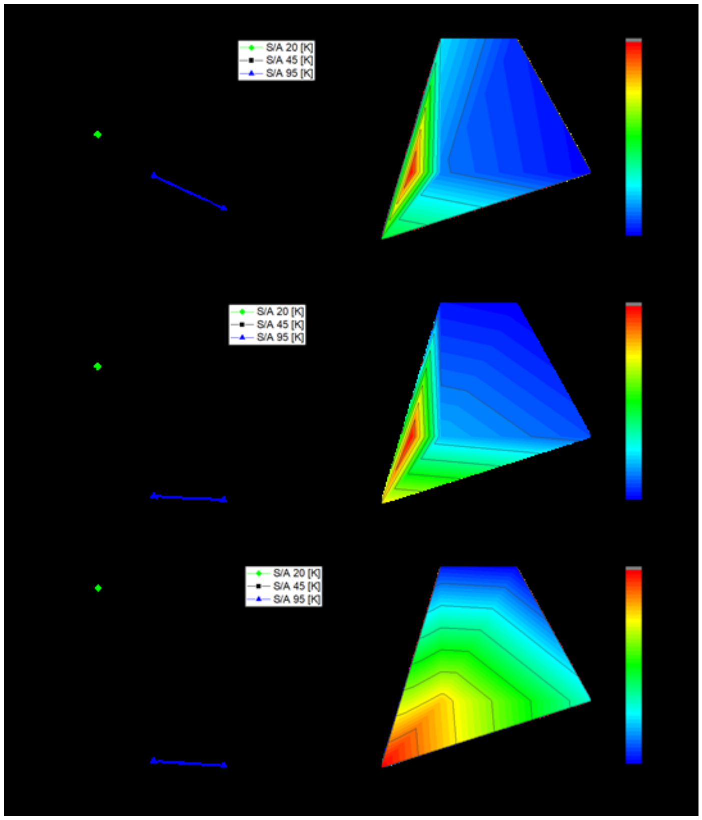


Figure 3. Linear plots and contour maps showing the temperature and laser pulse energy depends of ion ratios with different charge states in the solid solution. A1-A2) Nb^{3+}/Nb^{2+} ratio, B1-B2) Zr^{3+}/Zr^{2+} ratio, and C1-C2) Nb_{total}/total ratio, which $Nb_{total} = Nb^{3+} + Nb^{2+}$ and total = $Nb^{3+} + Nb^{2+} + Zr^{3+} + Zr^{2+}$.

Dr. Bachhav's work advanced the state of the art and facilitated in-depth understanding of the APT instrument and methodologies associated with generation and detection of the ions from the sample. The work has resulted in three publications for FY 2020. Those publications are given below.

1. "Effect of proton pre-irradiation on corrosion of Zr-0.5 Nb model alloys with different Nb distributions," Z Yu, T Kim, M Bachhav, X Liu, L He, A Couet. Corrosion Science, 108790
2. "Microstructure and fission products in the UCO kernel of an AGR-1 TRISO fuel particle after post irradiation safety testing," Z Fu, IJ van Rooyen, M Bachhav, Y Yang. Journal of Nuclear Materials 528, 151884.
3. "A transmission electron microscopy study of EBR-II neutron-irradiated austenitic stainless steel 304 and nickel-base alloy X-750," X Liu, L He, H Yan, M Bachhav, JF Stubbins Journal of Nuclear Materials 528, 151851.

1.3 Dr. Fidelma Di Lemma

Dr. Fidelma Di Lemma has continued to refine sample preparation methods for electron backscatter diffraction (EBSD) on reactive irradiated materials (0.2 FTE). EBSD is an analytical technique to evaluate the microstructure of a specimen (including grain size, texture and orientation, and deformation and stresses. The quality of the data obtained depends essentially on the quality of the sample surface, as the EBSD patterns are generated within a depth of less than 50-100 nm in the sample. Because of this, the data quality is extremely sensitive to the integrity of the crystallographic lattice order at the surface of the sample and to oxidation.

Dr. Di Lemma prepared a white paper entitled "EBSD analyses on metallic fuel samples: tips and improvements in sample preparation" which is attached as Appendix A. This document discusses the needs of sample preparation techniques for Al, Zr, U, U-alloys (Zr/Mo), Pu and alloys and irradiated fuel samples.

Based on her progress in sample preparation during FY 2020, Dr. Di Lemma applied EBSD to the understanding of microstructural evolution that result from fabrication processes, after transient nuclear events, and steady-state in pile irradiations. Dr. Di Lemma selected representative material from each of the three categories, and applied EBSD to evaluate the grain structure.

In FY 2020, Dr. Dilemma was able to markedly improve the EBSD characterization for texture analyses, provided first of a kind results of EBSD on irradiated fuel samples, improved the application of EBSD to a wide range of metallic fresh fuel samples, and improved the use of EBSD on cladding materials, in particular, difficult to analyze aluminum and zirconium based alloys.

This research was instrumental in improving the more routine application of EBSD at INL. The funding provided by NSUF IS has enabled more comprehensive analysis of a wide range of samples and cladding materials. The EBSD methods developed have been used to support NSUF rapid turnaround experiments (RTE) and has enabled the development of new research areas submitted as proposals submitted to NSUF and other areas. The NSUF IS support has also been used to prepare (3) and submit (2) journal articles, listed below.

1. Fidelma G.Di Lemma, LiuXiang, Tommy V.HolschuhII, Charles P.Folsom, Daniel J.Murray, TengFei, Colby B.Jensen; "Investigation of the microstructure evolution of alpha uranium after in pile transient," Journal of Nuclear Materials, Vol 542. Dec 2020.
2. F.G. Di Lemma, J.Burns, J.W.Madden, A.J.Winston, A.B.Robinson, J.F.Jue, D.D.Keiser, J.I.Cole; "Texture analyses and microstructural evolution in monolithic U-Mo nuclear fuel," Journal of Nuclear Materials, Vol 541, Nov 2020.
3. "Additive manufactured ODS Steel for FCCI mitigation," which is in preparation.

1.4 Dr. Lingfeng He

Dr. Lingfeng He is a distinguished staff scientist and leads the TEM (Transmission Electron Microscopy) group. He is a subject matter expert in the application of the TEM to fuels, reactor core materials, and fundamental understanding of fission gas bubble formation in irradiated fuels. Dr. He was supported for 0.2 FTE during FY 2020.

The technical achievements accomplished by Dr. He utilizing the NSUF IS funding are briefly summarized below:

1. Phase and defect evolution under irradiation in U-N-O. Although nitride-based fuels are widely considered as nuclear fuels, this is the 1^{st} detailed study defect evolution, resulting in a manuscript under review by Acta Mater, and another one is under preparation.
2. Atomic resolution characterization of fission gas bubbles (Figure 4) and dislocations in UO_2 coupled with advanced modeling to determine the gas bubble pressure. This research adds critical information to the understanding of fission gas behavior, often life limiting in nuclear fuels. A manuscript is under preparation.
3. Electron energy loss spectroscopy (EELS) data analysis using principal component analysis (PCA) in the HyperSpy analysis package was developed for removing noise from EELS (Elecgtron Energy Loss) spectra. A manuscript has been prepared and will be submitted.
4. Developed a quantification method using EELS for minor element (e.g., Ce) additions in oxide materials.

A highlight from the state-of-the-art work performed on the TEM utilizing NSUF IS funding is shown in Figure 4, an atomic resolution STEM image of an intergranular bubble (Kr) in irradiated UO_2. Each dot corresponds to a U atom column. Despite the importance of the behavior of fission gas in fuels, a comprehensive atomic level understanding of the evolution of fission gas bubbles in fuels and their interaction with the microstructure is still lacking. Such a fundamental understanding will enable the design of high burn-up fuels by making possible the prediction of the thermal and mechanical behavior of these fuels under reactor conditions. In this regard, characterizing fission gases in the fuel at atomic scale is important.

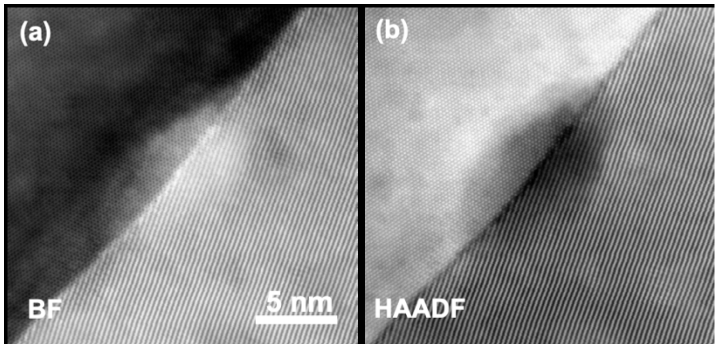


Figure 4. Atomic resolution scanning transmission electron microscopy (STEM) image of an intergranular krypton (Kr) bubble in irradiated UO_2. The electron beam is along the [011] direction for the top left grain. Each dot corresponding to a U atom column. (a) Bright field (BF) and (b) high angle annular dark field (HAADF) images

The publications resulting from the NSUF IS funding are summarized below. The NSUF IS funding contribution to the work is summarized at the end of each publication citation. In addition to the publications, NSUF IS funding partially supported (through time charged) attendance at the TMS conference, where 13 presentations were given, and the MRS fall meeting (3 presentations).

1. S. Johns, **L. He**, K. Bustillo, W.E. Windes, R. Ubic, C. Karthik: Fullerence-Like Defects in High-Temperature Neutron-Irradiated Nuclear Graphite, ***Carbon***, 166 (2020) 113-122Y.
2. Wang, T. Yao, F. Xi, P. Lei, X. Guo, **L. He**, G. Frankel, J. Lian: Degradation Mechanisms of Lead-Vanado-iodoapatite in Liquid-mediated NaCl Solutions, ***Corr. Sci.*** 172 (2020) 108720. (*Microstructural characterization using STEM/EDS and data analysis was supported by NSUF-IS*).
3. Y. Zhang, Y. Lin, **L. He**, M. Vijayakumar, M.S. Bhuvaneswari, G. Pawar, B.M. Sivakumar, H. Ding, D. Ding, B. Liaw, E.J. Dufek, Bin Li: Dual Functional Ni_3S_2@Ni Core-shell Nanoparticles Decorating Nanoporous Carbon as Cathode Scaffolds for Lithium-Sulfur Battery with Lean Electrolytes, ***ACS App. Energy Mater.*** 3 (2020) 4173-4179. (*Microstructural characterization using STEM/EDS and data analysis was supported by NSUF-IS*).

1.5 Dr. Scott Middlemas

Dr. Scott Middlemas is an Instrument Scientist specializing in the measurement of thermal properties. He was funded at 0.2 FTE for training and research centered on the development of the thermal conductivity microscope (TCM). The TCM is now installed in the Thermal Properties Cell in the Irradiated Materials Characterization Laboratory (IMCL). Research by Dr. Middlemas in FY 2020 focused on the demonstration and refinement of the “point and shoot” mode of the TCM. This work resulted in the measurement of spatially resolved thermal conductivity and thermal diffusivity from an irradiated multiphase U_3Si_2 fuel sample.

The microstructure of the U_3Si_2 fuel plate was mapped as shown in Figure 5. Figure 5A to Figure 5F show enlarged sections of a scanning electron microscopy (SEM) map containing the locations at which thermal conductivity and thermal diffusivity measurements were performed. The SEM images show the fuel particles in light colors consistent with the presence of the heavy element uranium. The dark regions represent the aluminum matrix in which the particles are embedded.

The measurements of local thermal diffusivity and thermal conductivity are presented alongside local porosity and fission density in Table 3. The locations A to F in Table 3 correspond to Figure 5A to Figure 5F. The specific heat values have been derived from the thermal conductivity, thermal diffusivity, and density (obtained by correcting for local porosity). The local porosity was estimated using an algorithm to analyze back scattered electron images. The measured thermal conductivity was modeled using a solid state physics model that includes the effects of porosity; results are shown in Figure 6A. Porosity in the fuel is driven by fission-related processes, and Figure 6B shows thermal conductivity as a function of fission density.

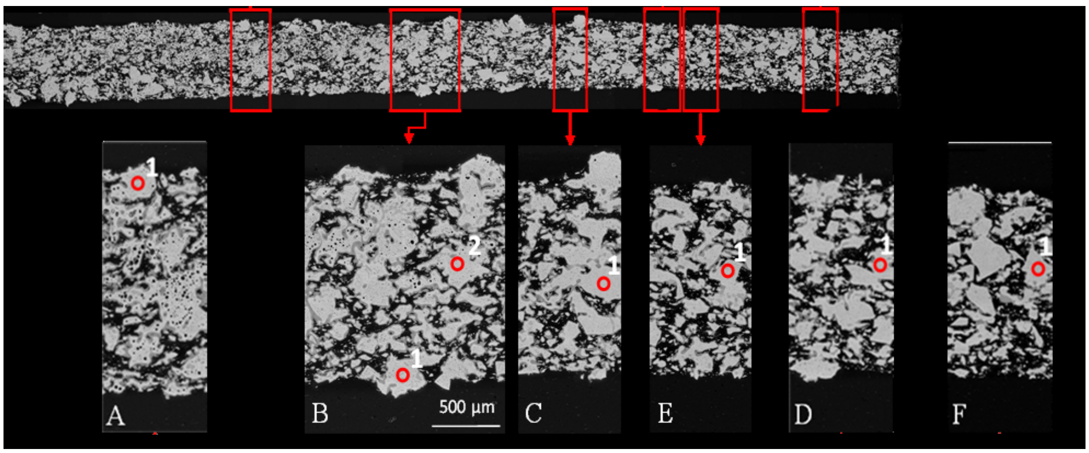


Figure 5. A secondary electron image of a cross section of U_3Si_2 fuel plate. Sections (A) to (F) show the locations at which local thermal conductivity measurements were performed.

Table 3. Table of measured properties and characteristics for each of the locations in Figure 5.

location	fission density (fissions cm^{-3})	specific heat (J kg^{-1} K^{-1})	thermal diffusivity (mm^2 s^{-1})	diffusivity error (mm^2 s^{-1})	porosity (-)	thermal cond. (W m^{-1} K^{-1})	conductivity error (W m^{-1} K^{-1})
A1	4.923	167.3	3.080	0.232	0.114	5.615	0.784
B1	4.328	179.1	3.270	0.344	0.100	6.480	0.758
B2	4.251	148.8	3.530	0.244	0.093	5.860	0.423
C1	4.007	156.5	3.207	0.254	0.110	5.495	0.251
D1	3.928	168.8	3.437	0.330	0.101	6.416	0.373
E1	3.831	155.6	3.409	0.406	0.110	5.808	1.166
F1	3.607	172.4	3.270	0.202	0.119	6.110	0.480

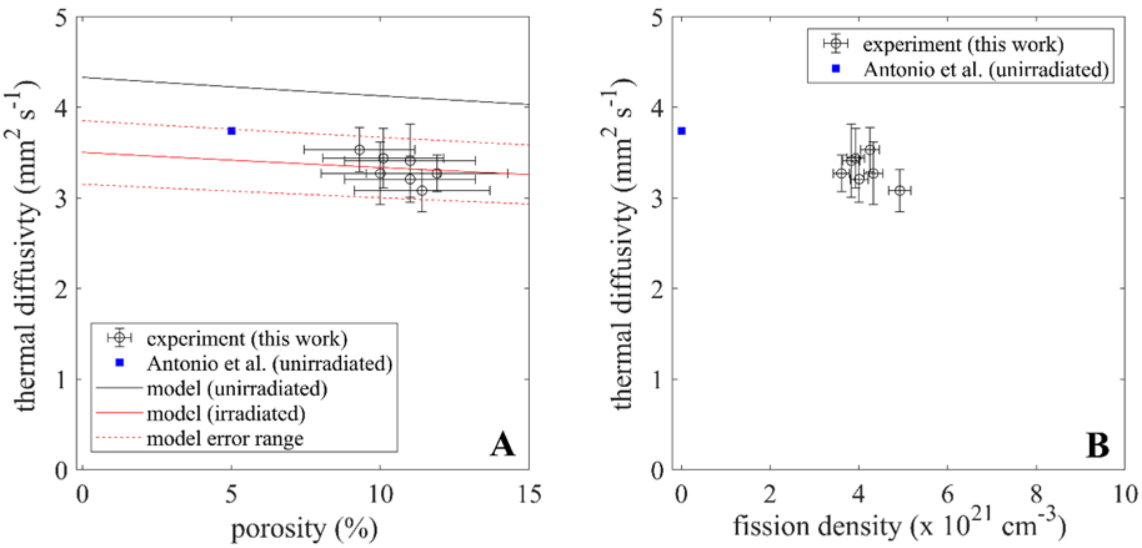


Figure 6. Local thermal conductivity measurements of the irradiated U_3Si_2 fuel particles at room temperature. A) Thermal conductivity plotted as a function of porosity. The irradiated measurements in this work are compared to literature data on fresh U_3Si_2 (J.T. White, A.T. Nelson, J.T. Dunwoody, D.D. Byler, D.J. Safarik, K.J. McClellan, Thermophysical properties of U3Si2 to 1773K, Journal of Nuclear Materials, 464 (2015) 275-280 and D.J. Antonio, K. Shrestha, J.M. Harp, C.A. Adkins, Y. Zhang, J. Carmack, K. Gofryk, Thermal and transport properties of U3Si2, Journal of Nuclear Materials, 508 (2018) 154-158.) and to the model prediction. B) Thermal conductivity plotted as a function of fission density.

This work measured, for the first time, the evolution of thermal conductivity as a function of microstructural and compositional changes caused by fission. It is evident that thermal conductivity decreases with increasing fission density. These results are explained with a solid states physics model and compared to literature values in a publication to be completed and submitted in the next FY.

Dr. Middlemas's work has resulted in 1 publication accepted (work performed last FY) and one publication in press. In addition, Dr. Middlemas presented a presentation at the M&M Conference in Milwaukee, WI in June of 2020 (virtual).

1.6 Dr. Daniel Murray

Dr. Daniel Murray is an early career instrument scientist specializing in the use of focused ion beam (FIB) instruments. Dr. Murray was funded at 0.2 FTE for FY 2020. Dr. Murray has focused his efforts on using the FIB platform to facilitate site specific measurement of the electronic and thermophysical properties of materials using FIB methods coupled with the PPMS (Physical Property Measurement System).

Accurate measurement of physical properties typically requires the growth of a single crystal, which is logistically difficult for actinide materials of interest (Np, Pu, Am) and nearly impossible for some material systems because of thermodynamic constraints. The dual beam scanning electron/focused ion beam microscope (FIB/SEM) is a powerful characterization platform that offers the ability to extract and manipulate extremely small selcted areas of bulk samples, deposit conductive materials, enables detailed phase identification, grain size and orientation measurements, and microchemical characterization of materials. While these instruments are extremely powerful in their ability to characterize the microstructure of materials, they currently lack the ability to probe electrical and thermophysical proberties of materials.

Dr. Murray's research addresses this shortcoming by integrating the newly developed 3ω technique with the FIB platform to allow for characterization of the thermal and electronic properties of materials at the microscale while directly relating them to the microstructure of the material. The FIB enables the selection, microstructural and chemical characterization, orientation, extraction, and mounting of small single crystals from polycrystalline/multiphase material. This method eliminates the need for bulk single crystal specimens greatly expanding the number and rate at which the properties of actinide materials can be measured, and expanding measurements to a much broader material set. This work has successfully demonstrated the integration of the 3ω method to study the thermal properties of micro-size samples that will play an important role in understanding the thermal conductivity of materials, especially those where phonon boundary scattering dominates thermal resistance, including nuclear materials and technologies such as microelectronics and thermoelectric materials.

These methods are being developed for the study of actinide materials and nuclear fuels on the microscale, where the effect of strong electronic correlations in 5*f*-electron orbitals result in unusual properties. To demonstrate this technique, we studied thermal transport in a microscale SmB_6 sample (Figure 7a). Using the Wiedemann-Franz law, we were able to separate the electronic, $\kappa_e(T)$, and the phonon, $\kappa_{ph}(T)$, contributions to the total thermal conductivity (Figure 7b). Further, we used the Calloway model to obtain the temperature dependence of $\kappa_{ph}(T)$. In contrast to standard behavior non-magnetic insulators, where phonon-phonon interactions lead to a reduction of the thermal conductivity with an increase of temperature, we find that the thermal conductivity increases with increasing temperature, creating a maximum at around 300K for micro crystals. This points to complicated phonon interactions in this topological material that are characteristic of resonant scattering.

To apply this work to the measurement for the electronic and thermo-physical properties of pure actinide materials, we have started development of a method for obtaining thin films of isotopically pure elements. Using the mass separator located in the Analytical Research Laboratory at the Materials and Fuels Complex (MFC), we have deposited thin films of Sm-154 onto a silicon wafer (Figure 8). One big question using this method for thin film deposition is whether mixing of the deposited ions and the substrate occurs. To answer this, we performed SEM/FIB and EDS analysis. From the SEM imaging, we can see that the surface roughness of the deposited area is similar to the base substrate, indicating that sputtering was minimal and the main process occurring is deposition. This will further be analyzed using SIMS depth profiling to quantify the extent of mixing. Once the process has been verified to produce a thin film, we will use FIB and 3ω methods to measure the electronic and thermo-physical properties of isotopically pure elements.

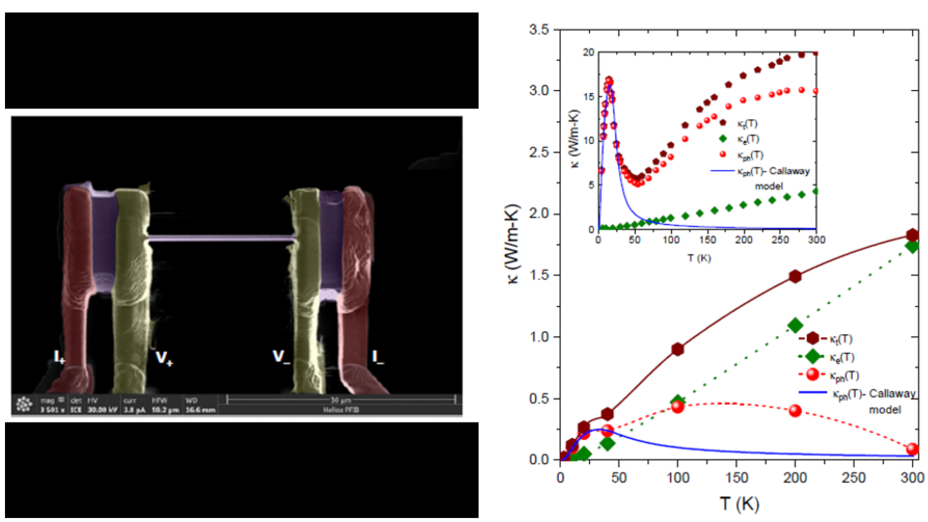


Figure 7. a) SEM image of a micro scale SmB6 sample with deposited electrical contacts in a 4-wire configuration. b) Thermal conductivity of the sample in A as a function of temperature. The $\kappa_e(T)$ and $\kappa_{ph}(T)$ represent the electronic and phonon contributions respectively.

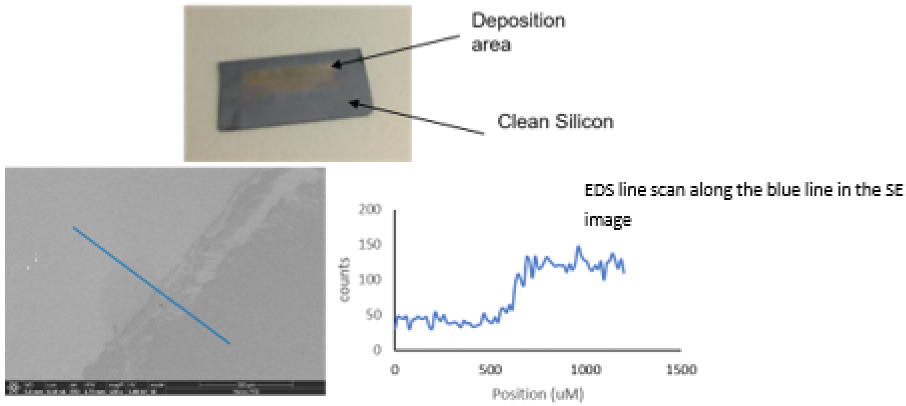


Figure 8. Deposition and SEM analysis of Sm-154 deposited onto a Si(100) wafer.

Dr. Murray has written one manuscript that has been accepted for publication and has submitted two publications this fiscal year, as summarized below.

1. M. Mofazzel Hosen, Gyanendra Dhakal, Baokai Wang, Narayan Poudel, Bahadur Singh, Klauss Dimitri, Firoza Kabir, Christopher Sims, Sabin Regmi, William Neff, Anan Bari Sarkar, Amit Agarwal, Daniel Murray, Franziska Weickert, Krzysztof Gofryk, Orest Pavlosiuk, Piotr Wiśniewski, Dariusz Kaczorowski, Arun Bansil & Madhab Neupane "Observation of gapped state in rare-earth monopnictide," Nat. Comm., *Sci Rep* **10,** 12961 (2020).
2. Poudel, D. Murray, J. Jeffries, and K. Gofryk, "Boundary scattering in micro-size crystal of topological Kondo insulator SmB_6" Applied Physics Letters, submitted.
3. Murray, D. J., Poudel, N., Gofryk, K., Review of Scientific Instruments, in preparation.

1.7 Dr. Xiang Liu

Dr. Xiang Liu performed work during FY 2020 on advanced TEM data acquisition and analysis using the Hyperspy software platform for advanced processing of TEM data, including EELS (Electron Energy Loss Spectroscopy) and EDS (Energy Dispersive Spectroscopy) spectral images and precision electron diffraction. Dr. Liu was supported at 0.2 FTE for FY 2020.

The application of the Hyperspy software has been used on numerous TEM data sets to enhance and quantify the results from TEM analysis. In particular, Dr. Liu used phase mapping using HyperSpy non-negative matrix factorization. This is illustrated in Figure 9 below.

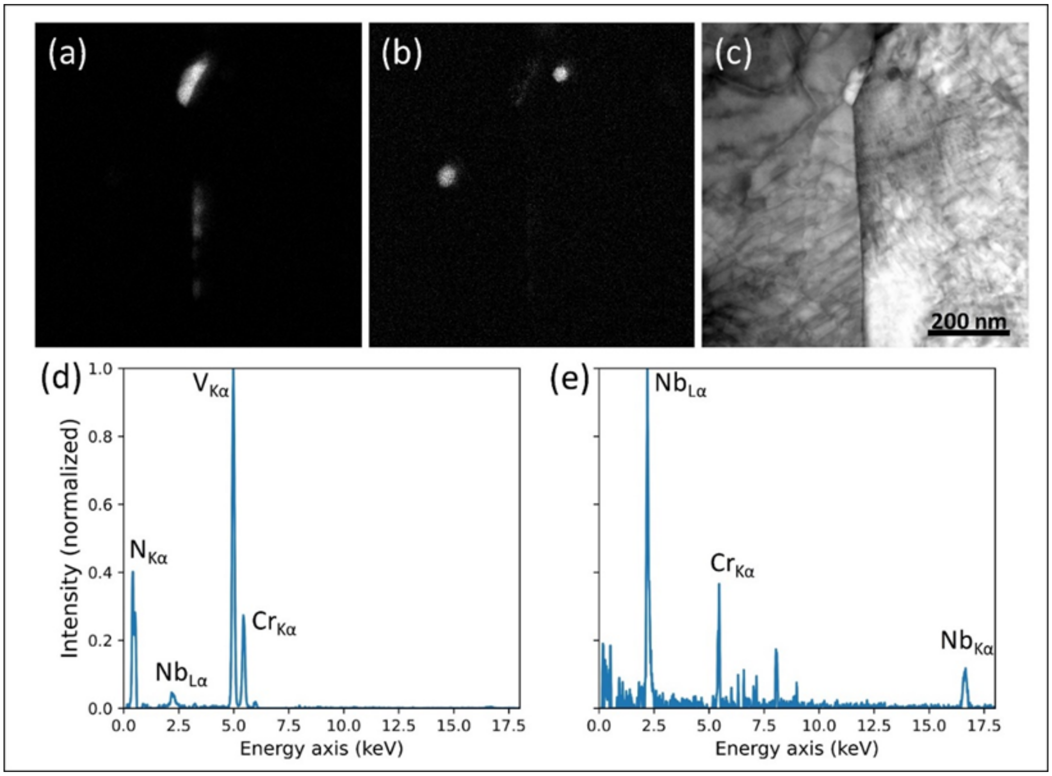


Figure 9. (a) V-rich nitride on grain boundary. (b) Nb-rich particles. (c) STEM-BF image. (d) spectrum of V-rich particles. (e) spectrum of Nb-rich particles. (a), (b), (d) and (e) are acquired from non-negative matrix factorization of EDS spectral image.

In addition to utilizing the hyperspy software, Dr. Liu also used the EELS system to identify a possible suboxide layer in a Zr-Nb alloy. Using the EELS effectively enhances the detection of light elements, allowing for better TEM imaging. Dr. Liu also was able to improve his expertise in precession electron diffraction (ASTAR) orientation mapping. The small grains (lath martensite structure) in an ODS (Oxide Dispersion Strengthened) alloy were well resolved using the ASTAR system. Conventional EBSD measurements in SEM cannot resolve these features.

Dr. Liu published one journal article using NSUF IS funding and has another under review. A summary of the publications is given below.

1. **Xiang Liu**, Lingfeng He, Huan Yan, Mukesh Bachhav, James F Stubbins, "A transmission electron microscopy study of EBR-II neutron-irradiated austenitic stainless steel 304 and nickel-base alloy X-750," Journal of Nuclear Materials 528 (2020) 151851.
2. Fidelma G. Di Lemma, **Xiang Liu**, Tommy V. Holschuh II, Charles P.Folsom, Daniel J. Murray, Fei Teng, Colby B. Jensen. "Investigation of the microstructure evolution of alpha uranium after in pile transient," Journal of Nuclear Materials 542 (2020) 152467.
3. **Xiang Liu**, Luca Capriotti, Tiankai Yao, Jason M. Harp, Michael T. Benson, Yachun Wang, Fei Teng, Lingfeng He, "Fuel-cladding chemical interaction of a prototype annular U-10Zr fuel with Fe-12Cr ferritic/martensitic HT-9 cladding," Journal of Nuclear Materials, Under Review.

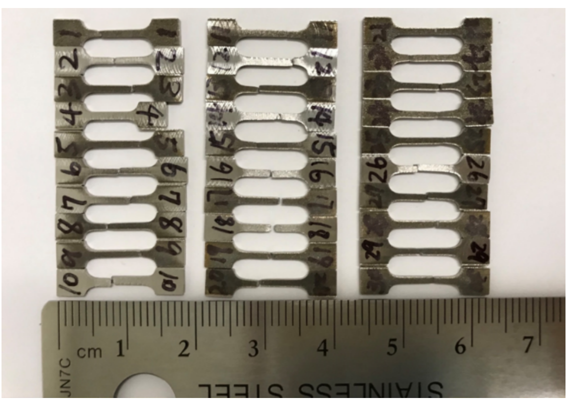
1.8 Dr. Cheng Sun

Dr. Cheng Sun is a senior staff scientist currently supported at 0.2 FTE. Dr. Sun's primary areas of NSUF research during FY 2020 was the development of a remote system for testing of miniature SS-J3 specimens typically used in NSUF irradiation experiments. SS-J3 denotes small specimens with J3 dimensions (16 mm x 4mm). The load frame was modified for use in IMCL shielded cells. Testing in compression and tension modes indicated acceptable measurement repeatability, as indicated in Figure 10.

In addition to developing the load frame, Dr. Sun investigated the feedstock of 316 stainless steel powders used in additive manufacturing and the nanoporous microstructure of printed 316L stainless steel specimens. Analysis included strain mapping around the pore structure. The mechanical properties of various 316 stainless steels (wrought, additive manufactured 316Lp and additive manufactured 316Lv) were measured. Deformation-induced twins and martensitic phase transformation were observed. The martensitic phase transformation was found to preferentially occur around the pore structure.

Dr. Sun wrote and submitted two journal articles using IS funding, listed below.

1. C. H. Shiau, M. D. McMurtrey, R. C. O'Brien, N. D. Jerred, R. D. Scott, J. Lu, X. Zhang, Y. Wang, L. Shao, **C. Sun***, "Deformation behavior and irradiation tolerance of additively manufactured 316L stainless steel," Materials & Design, submitted.
2. **C. Sun***, Y. Wang, M. D. McMurtrey, N. D. Jerred, F. Liou, J. Li., "Additive Manufacturing for Energy: A Review," Applied Energy, submitted.



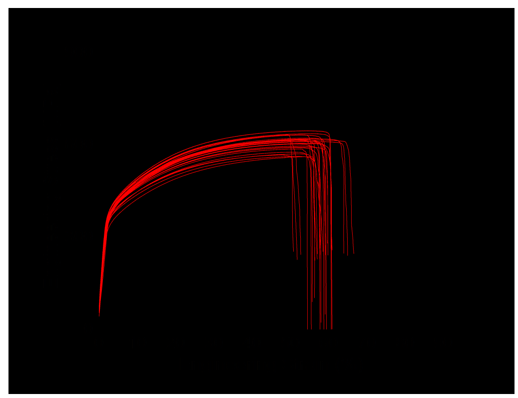


Figure 10. Repeatability testing on the load frame for miniature testing. Test for sample #4 was stopped due to misalignment.

1.9 Dr. Fabiola Cappia

Dr. Fabiola Cappia is a staff scientist who was supported at 0.2 FTE level to perform research on increasing the effectiveness of microscopy by developing applied methods for nuclear material microstructure characterization and 3D reconstruction during FY 2020. However, due to programmatic conflicts and the COVID-19 complications, she was only able to perform limited work on the topic this year. Dr. Cappia completed two publications which are listed below.

1. R. Genoni, D. Pizzocri, F. Antonello, T. Barani, L. Luzzi, T. R. Pavlov and F. Cappia*, "3D reconstruction of two-phase random heterogeneous material from 2D sections: An approach via genetic algorithms," JNM, in review.

2. R. Genoni, D. Pizzocri, F. Antonello, T. Barani, L. Luzzi, T. R. Pavlov and F. Cappia*, "Three-dimensional reconstruction from experimental two-dimensional images: application to irradiated metallic fuel," JNM, in review.

1.10 Dr. Aaron Craft

Dr. Aaron Craft participated in the NSUF Instrument Scientist Program during FY 2020 (0.2 FTE) with a focus on increasing the information that can be extracted and expand the application space of neutron imaging to serve the needs of nuclear research community, particularly for NSUF funded research.

Multi-Energy Neutron Imaging: Neutron imaging typically uses thermal (low energy) neutrons, but epithermal neutron radiography is also feasible at the Neutron Radiography Reactor (NRAD). Epithermal neutrons offer higher penetration and lower contrast than thermal neutrons, and, therefore, are able to radiograph some objects that cannot be radiographed with thermal neutrons. Epithermal and thermal neutron computed tomography, i.e., multi-energy nCT, can be combined to potentially extract additional information.

Building on demonstration of multi-energy imaging in FY 2019 [*Schillinger & Craft, Materials Research Proceedings 15 (2020) 142-148*], both epithermal & thermal nCT (neutron Computed tomography) first-of-a-kind datasets were acquired from the SETH-E Accident Tolerant Fuel (ATF) rodlet irradiated in the TREAT reactor. Combination, reconstruction, and analysis of the combined data sets is ongoing. The results of this work have already been invited for publication in the Journal of Imaging.

Radioactive Sample Stage: Work has been proceeding on developing a shielded sample holder capable of remotely positioning a sample into an x-ray and/or neutron beam and is equipped for transfers within a facility. A prototype is being built for use in IMCL's Zeiss Versa x-ray Microscope.

1.11 Dr. Nikolaus Cordes

Dr. Nikolaus Cordes joined the INL in December of 2018 as the IS for the x-ray microscope (XRM) (also known as or XCT, X-ray Computed Tomography). He was supported by NSUF at 0.2 FTE for FY 2020 to improve imaging and data collection from the Zeiss X-ray microcscope.

Dr. Cordes has been successful in a number of fronts. He continues to develop startegies for the characterizaiton of non-irradiated and slightly irradiated samples. He has worked to design a sample holder and work through health physics concerns for the handling samples and posting and downposting the instrument. The posting and downposting of the instrument is vital to the ease of use of the instrument to support all reasearch activities in IMCL. In addition, Dr. Cordes has worked to develop advanced sample processing methods to better display and provide more information to researchers. He also is actively working on developing collaborations that would allow the inclusion of the X-ray microscope to future NSUF supported work.

1.12 Drs. Leah Squires and Luiza Albuquerque

Drs. Leah Squires and Luiza Albuquerque have been conducting research and development on methods to produce pure Np metal and analytical techniques to characterize the purity level of the Np metal produced. The combined effort for this activity was 1 FTE. Dr. Squires worked to produce the metal Np (Figure 11) product through direct oxide reduction. During FY 2020, Dr. Squires produced approximately 11 grams of Np metal for use in INL programs and projects (in addition to the approximately 20 grams produced last year). Metallic Np can now be produced reliably when needed. .

Dr. Squires led the effort to provide a sample of the Np metal to Florida State University (FSU) for joint research. In January 2020, 0.888 grams of Np metal was packaged under Ar in double airtight containers. This was done to minimize the possibility of oxidation of the Np metal in transit. The sample was received without incident at FSU.

Dr. Albuquerque and her team were working to refine methods to provide the assay of the Np metal produced. Initially impurities in the metal (primarily Ta, Fe, U, Th, Ca and Pu) were identified and. quantified, assuming that the remainder of the metal can be assumed to be Np. The results are summarized in Table 4. Using this method, provides only a theoretical purity, and development of a method for analysis of Np within standard refence limits is ongoing.

Figure 11. Neptunium metal produced via calcium metal/calcium chloride reduction system.

Oxygen and nitrogen were determined using an Eltra ONH-2000 analyzer. Briefly, the sample is melted using a high temperature furnace. The oxygen is converted to CO_2 and measured in an IR-cell, while nitrogen is measured as N_2 using a thermoconductivity cell. Six ~ 0.1 g samples of the metal were cleaned using metal file and silicon carbide paper, sealed inside airtight containers inside of an inert atmosphere glovebox and transferred to the analyzer. The average concentration of nitrogen was measured to be 810 ppm (STD115 ppm). The average oxygen concentration was 547 ppm (STD 110 ppm).

1.13 Karen Wright

Karen Wright was supported via NSUF IS funding at the 0.2 FTE level for FY 2020. Because of major instrument reliability issues, Karen was unable to perform experiments leading to the development of improved methods for quantification of elements in a high Z matrix using the EPMA (Electron Probe MicroAnalysis) instrument. Ms. Wright was productive in writing two journal articles. One has been accepted for publication, and the other is in review. She also had one conference presentation supported in part by NSUF IS funds. The list of publications is given below.

1. **K. Wright** and I. Van Rooyen, “Fission Product Distribution in Irradiated Safety-Tested and Non-Safety- AGR2 TRISO Particles,” accepted IOP Materials and Science Engineering Conference Series.
2. **K. Wright**, J. Stempien, I. Van Rooyen, “Fission Product Distribution in Irradiated Safety-Tested and Non-Safety- AGR2 TRISO Particles,” submitted to JNM.

Table 4. HR-ICP-MS analysis results, including possible interferences and the RP used for successful separation. MQL is the Method Quantification Limit.

Analyte	*RP*	*isotope used*	*Interferences*	*Conc. (ug/g)*	*SD*	*MQL (ug/g)*
Al	4000	27Al	-	28.9	±6%	3
Cr	2500	52Cr	36Ar16O, 38Ar14N, 40Ar12C	60.7	±7%	0.3
Fe	2500	56Fe	40Ar16O	406	±6%	10

I	2500	127I	238U16O++	<1	N/A	1
Nb	300	93Nb	-	1.55	±8%	0.003
Ni	300	60Ni	-	19.6	±14%	0.8
Pb	300	208Pb	-	<0.1	N/A	0.1
Sn	300	118Sn	-	<0.2	N/A	0.2
Ta	300	181Ta	-	1920	±4%	0.1
Th	300	232Th	-	103	±0.3%	0.006
Tl	300	203Tl	-	<0.001	N/A	0.001
V	4000	51V	12C35Cl, 14N37Cl	1.68	±12%	0.02
Y	300	89Y	-	10.8	±2%	0.005
233m/z	2500	233Pa	-	4.56	±7%	0.7
234U	300	234U	-	9.53	±2%	0.001
235U	300	235U	-	2.74	±2%	0.001
236U	2500	236U	-	4.25	±10%	0.5
238U	300	238U	-	115	±2%	0.01
239Pu	300	239Pu	-	35.6	±2%	0.002
240Pu	300	240Pu	-	6.05	±2%	0.002
241Am	300	241Am	-	1.16	±14%	0.002
242Cm	300	242Cm	-	2.55	±4%	0.001
243Am	300	243Am	-	0.243	±10%	0.001
244Cm	300	244Cm	-	0.0837	±8%	0.001
Total impurities (ug/g)		**2735 ± 4%**	**maximum achievable purity (%) (Σ MQL)**		**99.998**	
Total Np purity (%)		**99.726**				

1.14 Dr. Chuting Tan

Postdoctoral research associate **Dr. Chuting Tan** developed a capability for digital positron annihilation spectroscopy (PAS), supported at 0.2 FTE.

The fully functional PAS system consists of one ^{22}Na source, two BaF_2 detectors, two HPGe detectors, a time spectrometer, an energy spectrometer, a high voltage power supply, and a preamp power supply assembled from several sources. The experimental set-up can be seen in Figure 12. The HPGe detectors were refurbished with funding from the NSUF IS program.

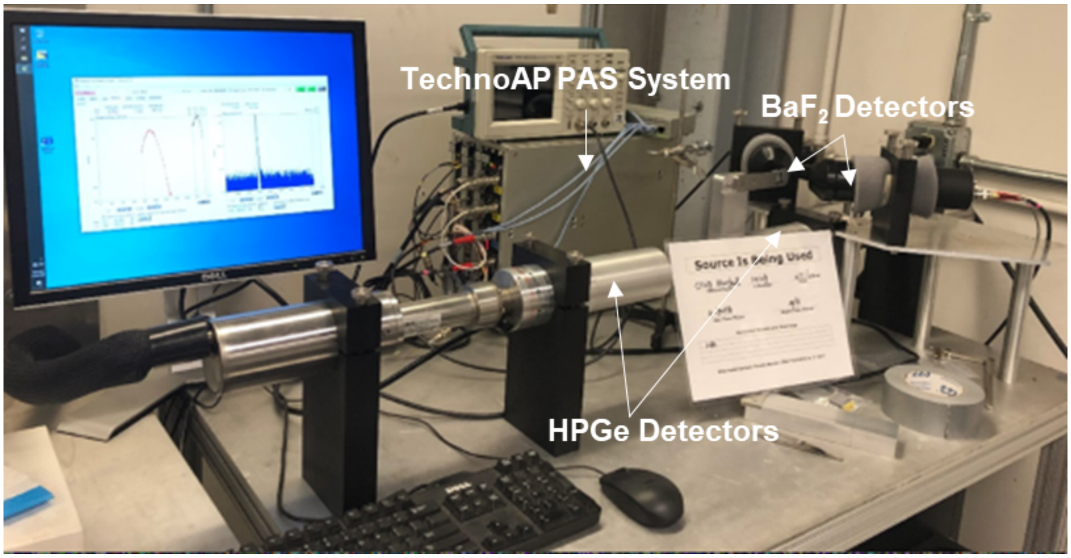


Figure 12. Full set up of the PAS system.

Measurements on several metals were conducted, including annealed aluminum, copper, and brass sheets. In collaboration with PAS scientists at INL, we acquired well characterized zeolite in silica gel and silicone (polydimethylsiloxane; PDMS) samples, which have long lifetime components (Figure 13). We benchmarked the measured lifetime results with those acquired by a similar instrument at INL and concluded that the system provides high quality measurements.

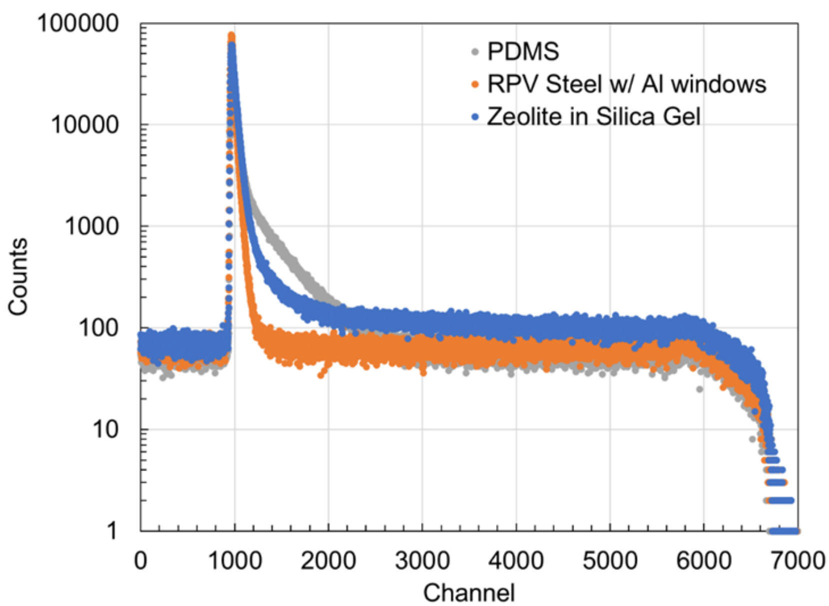


Figure 13. Lifetime spectra of several representative samples (RPV, PDMS, and Zeolite in silica gel

In addition to benchmarking the system setup, we also measured samples for other projects and programs with unknown lifetime, such as additively manufactured metal samples, irradiated reactor pressure vessel materials, and 3C-SiC material deposited by chemical vapor deposition method. All results were acceptable..

Based on the experience gathered during this IS project, Dr. Tan teamed up with two other PAS instrument scientists at INL, Jagoda Urban-Klaehn and Chase Taylor, on an LDRD proposal, “In-situ Positron Annihilation Spectroscopy for Characterizing Irradiation Induced Defects.” The proposal has been recommended for funding in FY 2021 and is currently in DOE review. The project seeks to further expand the PAS capability at INL investigating the feasibility of an in-situ positron beamline on one of the neutron sources (reactors) to track defect formation and. evolution in real time, a unique capability that may become available to NSUF in the future.

1.15 Dr. Nedim Cinbiz

Dr. Nedim Cinbiz, an early career researcher, was supported at 0.2 FTE for setting up and testing a digital imaging correlation (DIC) system to provide improved strain measurements for mechanical testing of irradiated materials.

NSUF IS funding supported the installation and the operation of Dantec stereo digital image correlation equipment and software. Stereo DIC is a non-contact metrology technique used to provide full-field strain mapping, resulting in improved understanding of material failure mechanisms. The DIC system will be integrated with the micromechanical testing stage being developed by Dr. Sun.

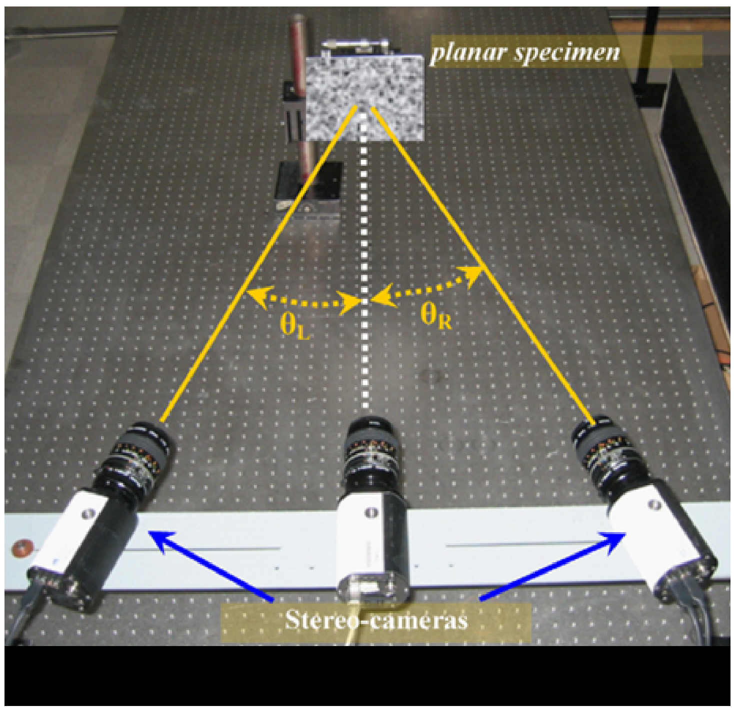


Figure 14. A typical stereo digital image correlation setup.

To evaluate the DIC software, previously collected data from Fe-based alloy have been used for the DIC analysis. Figure 15 shows a speckle-painted samples surface and imposed DIC results on that surface. DIC enables one to determine the two-dimensional strain distribution on the deformed specimen. Also, using specific DIC tools, line profiles or gauge strains can also be determined.

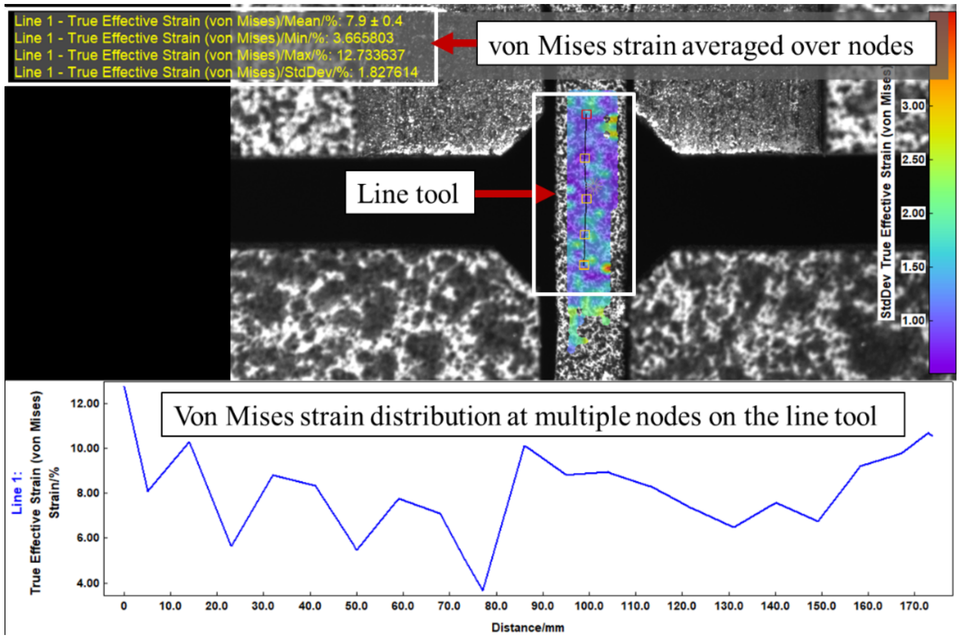


Figure 15. Typical DIC analysis on a deformed specimen surface

In addition, the NSUF IS funding supported the development of a non-contact technique for mechanical tests at INL. When fully implemented, this will eliminate the need to transport irradiated specimens to other laboratories for mechanical test characterizations.

1.16 Dr. Boopathy Kombaiah

Dr. Boopathy Kombaiah was assigned as the instrument scientist for the physical property measurement system (PPMS) for FY 2020, at the level of 0.2 FTE. Unfortunately, the PPMS system was out of service with a persistent cooling system problem during FY 2020. The cooling system was repaired as of August 2020, and the system is working to the manufacturer's specifications. An instrument scientist focused on the PPMS will be hired in FY 2021 to ensure that the system is used to its fullest potential.

1.17 Dr. Xiaofei Pu

Dr. Xiaofei Pu was the instrument scientist assigned to the X-ray Diffraction system during FY 2020. Xiaofei was supported at 0.2 FTE to become trained on the XRD instrument, oversee movement to a new location in IMCL, re-establish the XRD capability, and improve instrument capabilities.

Dr. Pu attended the Powder Diffraction and Quantitative X-ray Analysis Basics courses at the Bruker facility, where NSUF IS funded the time required to complete the training. Once trained, Dr. Pu was approved for independent work on the XRD instruments in IMCL. Dr. Pu continues to build interest among the INL programmatic staff for XRD analysis using the current instrument configuration and is developing a. user-informed plan for improving instrument capability. She also continues to build her skills and participated in virtual training on sample preparation and data analysis in September of 2020. In FY21, Dr. Pu will support NSUF RTE proposals, as well as implementing instrument improvements such as high temperature XRD capability.

2. INFRASTRUCTURE INVESTMENTS

The NSUF funded upgrades of the IMCL Titan TEM (adding an EELS and a probe corrector that will provide increased resolution to sub-angstrom levels (0.8Å) were completed in FY 2020. The upgrades have made the IMCL Titan TEM one of the most (if not the most) advanced TEM instruments to characterize irradiated fuel samples in the world.

3. CONCLUSIONS

The third year of the instrument scientist program has been successful. Each instrument scientist has made significant progress on their personal project, the development of their instrumentation and the improvement in the methods. These areas (summarized in Table 1) are important to either expand research in a particular strategic area, provide new capabilities to NSUF/INL, or to train instrument scientists on new instruments and methods. The instrument scientists were committed to publishing in peer-reviewed journals and presenting their results at national and international conferences.

One challenge encountered during FY 2020 was the impact of the COVID-19 epidemic on the ability of the IS to complete their work. Of notable importance was Ms. Karen Wright, Dr. Fabiola Cappia. In addition, several of the instruments suffered from significant outages as a result of delays in resolving instrument issues and upgrades, which impacted the overall IS program. Most significant were the Titan TEM, the PPMS instrument, and the EPMA instrument. The instrument problems along with the subsequent difficulty in obtaining vendor service associated with the virus complicated those work activities.

The instrument scientists attended the weekly NSUF staff meetings on a regular basis before the curtailment of work. As Dr. Kennedy suggested, the meetings were an ideal time for them to network with NSUF staff and to learn about the breadth of research funded through NSUF. The NSUF Instrument Scientist Program provided Instrument Scientists the opportunity to pursue development of capabilities that improve NSUF's scientific capability. These capabilities provide opportunities for discovery that often have a high impact on nuclear science and technology.