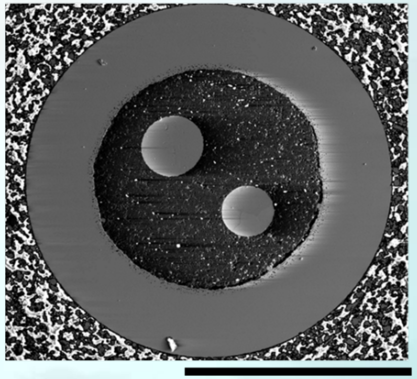
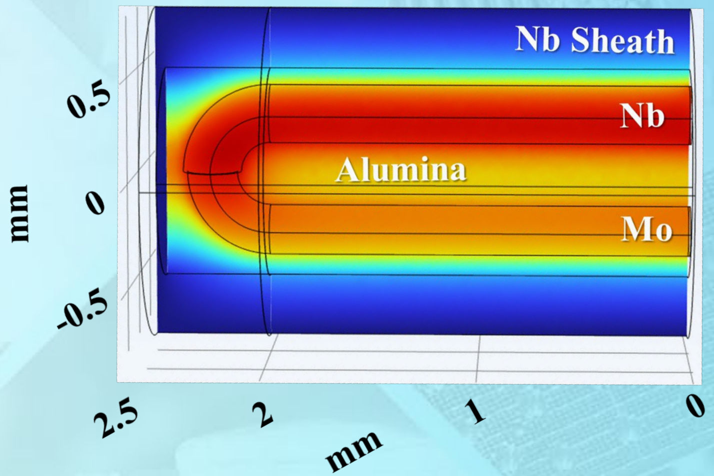
U.S. DEPARTMENT OF ENERGY | Office of NUCLEAR ENERGY

Advanced Sensors and Instrumentation

Nuclear Thermocouples





Advanced Sensors and Instrumentation (ASI)
Boise State University (BSU) Research Overview

April 13, 2023

BSU: Brian J. Jaques (PhD, PE) and Scott Riley (GRA)
INL: Richard Skifton
Boise State University (MSE) and Idaho National Laboratory

Project Overview

OVERVIEW

Purpose: The thermocouple element implements R&D activities to develop nuclear instrumentation that address critical technology gaps for monitoring and controlling existing and advanced reactors and supporting fuel cycle development. For temperature measurements, thermocouple instrumentation is typically composed of one or more sensing element, interrogation systems, data acquisition system as well as processes and procedures to collect, analyze and calibrate data. Instrumentation is used to measure process parameters, such as temperature, independently of the experiment, component or process in which it is deployed.

Objectives: The thermocouple activities:
- Characterize influence of traditional stabilization heat-treatment on HTIR-TCs
- Performance assessment of commercial TCs for nuclear applications
- Characterize influence of alternative heat-treating methods, Joule heating, on stabilization of HTIR-TCs

DETAILS

Principal Investigator: Brian Jaques (BSU), BrianJaques@BoiseState.edu
Richard Skifton (INL), richard.skifton@inl.gov

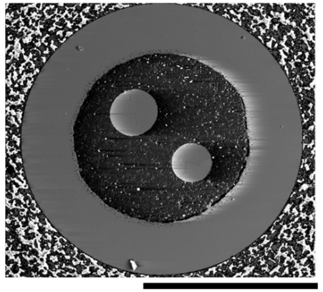
Institution: Boise State University

Collaborators: Idaho National Laboratory

Duration: FY2021-23

TPOC (Technical Point of Contact): Kort Bowman, kort.bowman@inl.gov

Federal Manager: Daniel Nichols



SEM micrograph of HTIR-TC cross-section

Schedule: The thermocouple activities:

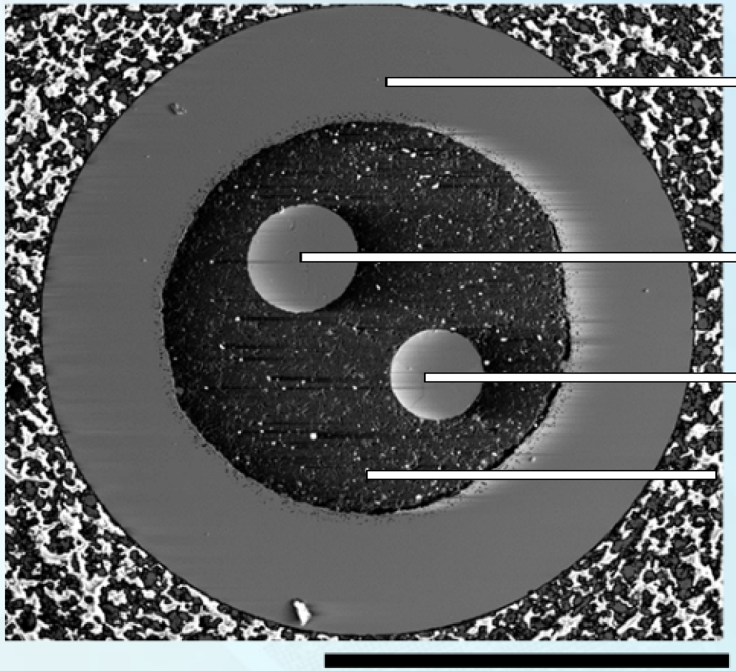
- ***Characterize influence of traditional stabilization heat-treatment on HTIR-TCs***
  - During the stabilization heat-treatment a secondary Nb_3P phase along with an alumina/Nb-P interaction region forms.
- ***Performance assessment of commercial TCs for nuclear applications***
  - A Halden-Conroy model was used to model the empirical drift due to transmutation for type C, S, and K.
- ***Characterize influence of alternative heat-treating methods, Joule heating, on stabilization of HTIR-TCs.***
  - A preliminary model using COMSOL was made to evaluate the temperature within the thermoelements during joule heating.

Technical Impact

- In order to decrease nuclear innovation time, robust, in-pile measurement techniques and sensors must be developed.

Gen IV: Very High Temperature Reactor Core Outlet Temperature[3]: >1000°C

Thermocouple[2]	HTIR-TC
Materials	Mo vs. Nb
Temp Range	0 – 1700 °C
Cost	~$250/ft
Radiation Tolerance as compared to HTIR-TC	

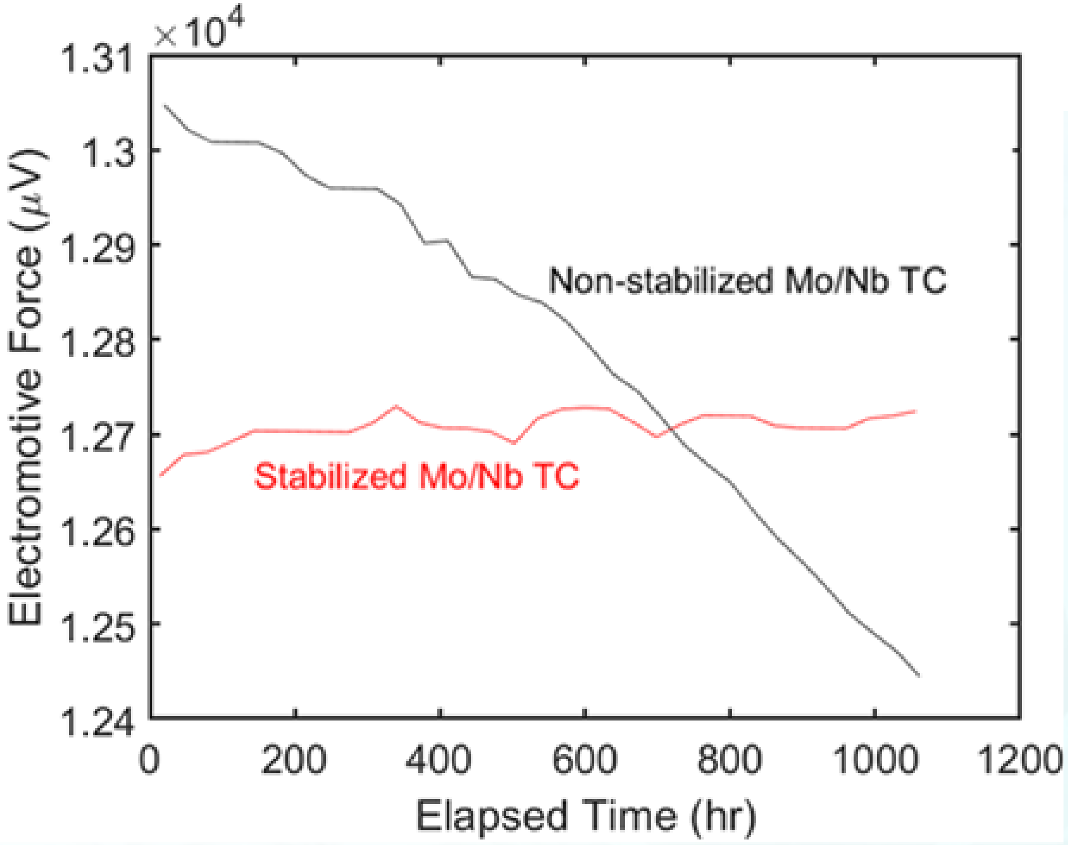


SEM micrograph of HTIR-TC cross-section

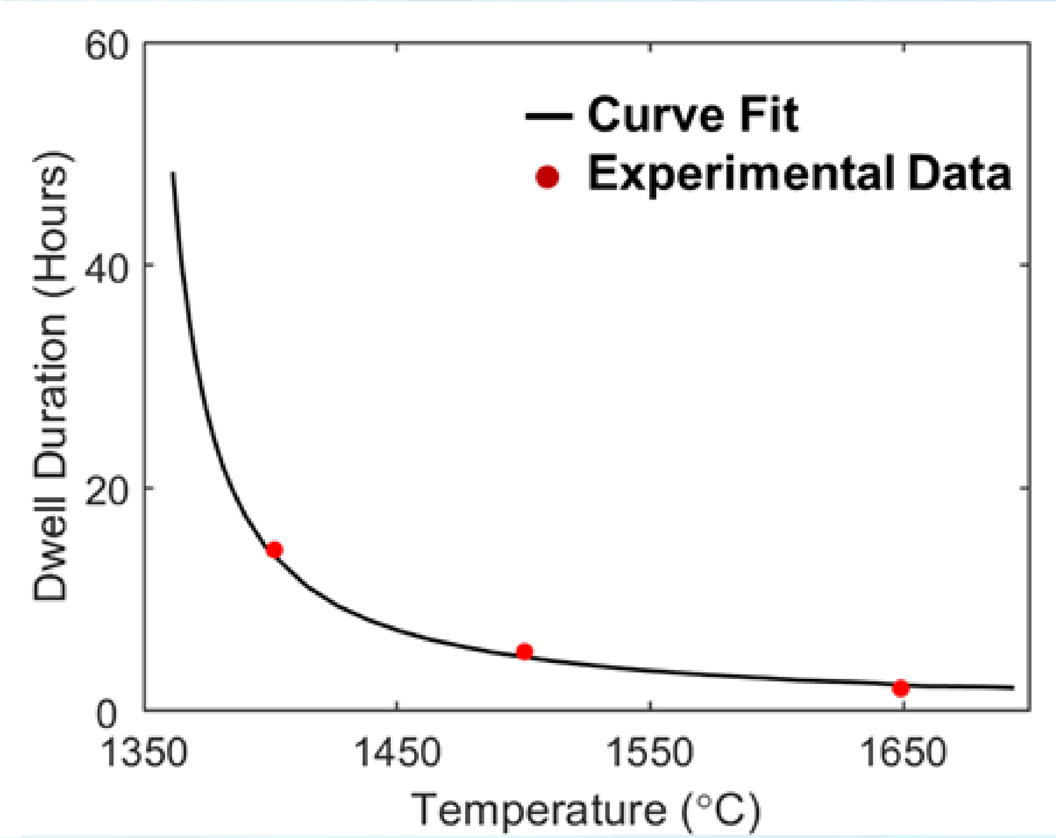
HTIR-TC combines the high temperature of the Type B thermocouple with the irradiation tolerance of Type N & K.

2. Data courtesy of Dr. Skifton, INL
3. Murty K., Charit I., An Introduction to Nuclear Materials. Vol 1, Wiley-VCH, 2013, Weinheim, Germany.

Technical Impact



Signal measured by Mo/Nb thermocouples during 1000-hour 1100 °C test [4].



Hours to stabilize as a function of heat treatment temperature for HTIR-TCs.

4. Bong Goo Kim, Joy L. Rempe, Jean-François Villard & Steinar Solstad (2011) Review Paper: Review of Instrumentation for Irradiation Testing of Nuclear Fuels and Materials, Nuclear Technology, 176:2, 155-187, DOI: 10.13182/NT11-A13294

Results and Accomplishments

Experimental:

- Investigated the effects of the stabilization heat treatment on the mechanical and electrical properties, chemical stability, and microstructural evolution of HTIR-TCs.
- Provided optimization procedures and suggestions for path forward for stabilization heat treatment strategies.
- Developed a thermocouple drift model based on the empirical data in open literature.

Presentations:

1. "Effect of Processing on Nuclear Thermocouple Stability for In-Pile Temperature Sensing." S. Riley, K. Holloway, R. Skifton, B.J. Jaques. Accepted for presentation at the International Conference on Advancements in Nuclear Instrumentation Measurement Methods and their Applications (ANIMMA). Real Collegio, Italy. June 12-16, 2023.
2. The Influence of Heat Treatment on the Stability of High Temperature Irradiation Resistant Thermocouples." S. Riley, K. Holloway, A. Bateman, R. Skifton, B.J. Jaques. Presented at the FuNZI 2023 Conference. March 30-31, 2023.
3. "High Temperature Irradiation Resistant Thermocouples for In-Pile Temperature Sensing." S. Riley, K. Holloway, R. Skifton, B.J. Jaques. Presented at the MS&T 2022 Conference. October 9-12, 2022.
4. "Performance of niobium and molybdenum alloys for high temperature sensing applications." S. Riley, B. Perrine, E. Sikorski, L. Li, R. Skifton, B.J. Jaques. Presented at the TMS 2020 conference. San Diego, CA. February 23-27, 2020.
5. "Development and performance of high temperature irradiation resistant thermocouples." S. Riley, B. Perrine, E. Sikorski, R. Skifton, L. Li, B.J. Jaques. Presented at the Materials Science and Technology 2019 Conference. Portland, OR. September 29-October 3, 2019. INVITED.

Other:

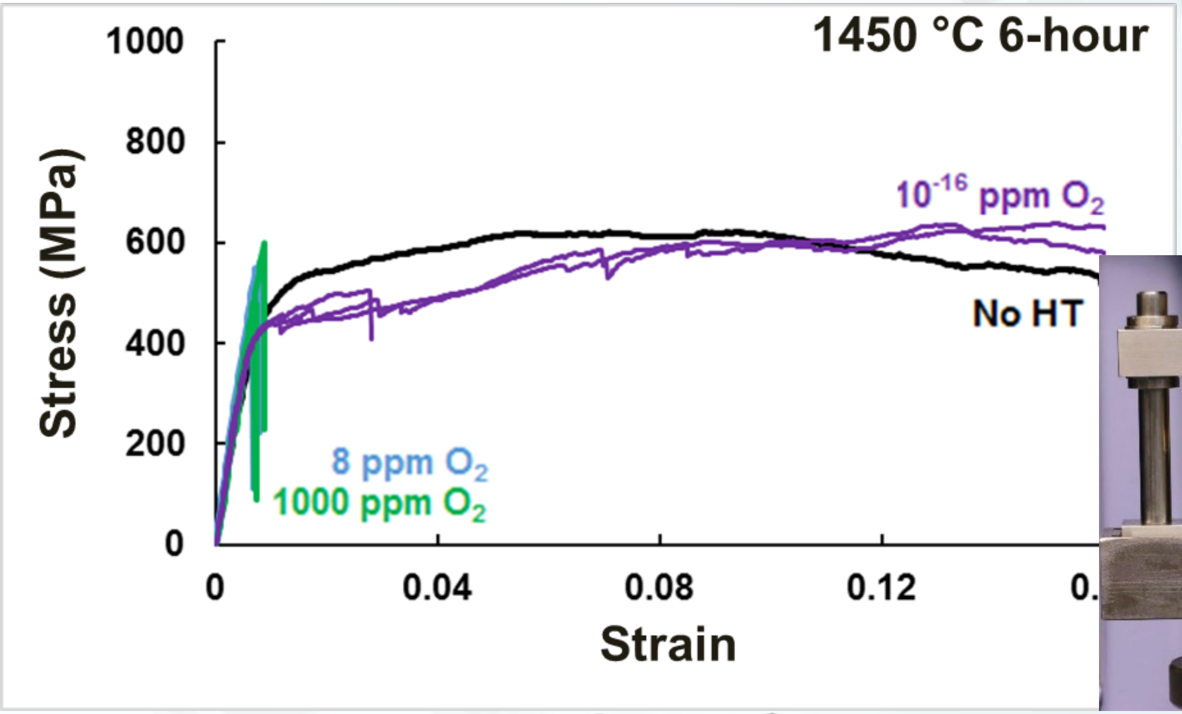
- MaCS Seed Grant proposal: $5k to develop mechanistic understanding of Mo thermoelement in HTIR-TCs, "Influence of heat treatment on the grain stability of Mo-LaO".
- Scott Riley (GRA) began internship on April 3rd, 2023 – optimization of HTIR-TC builds and stabilization heat treatment via Joule heating

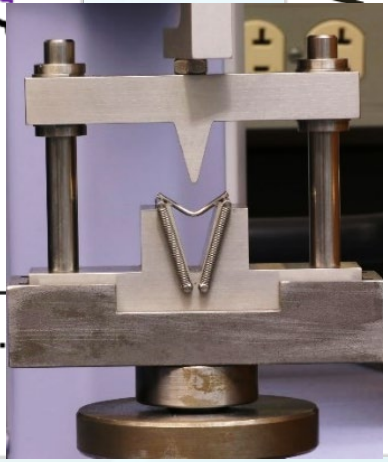
Publications:

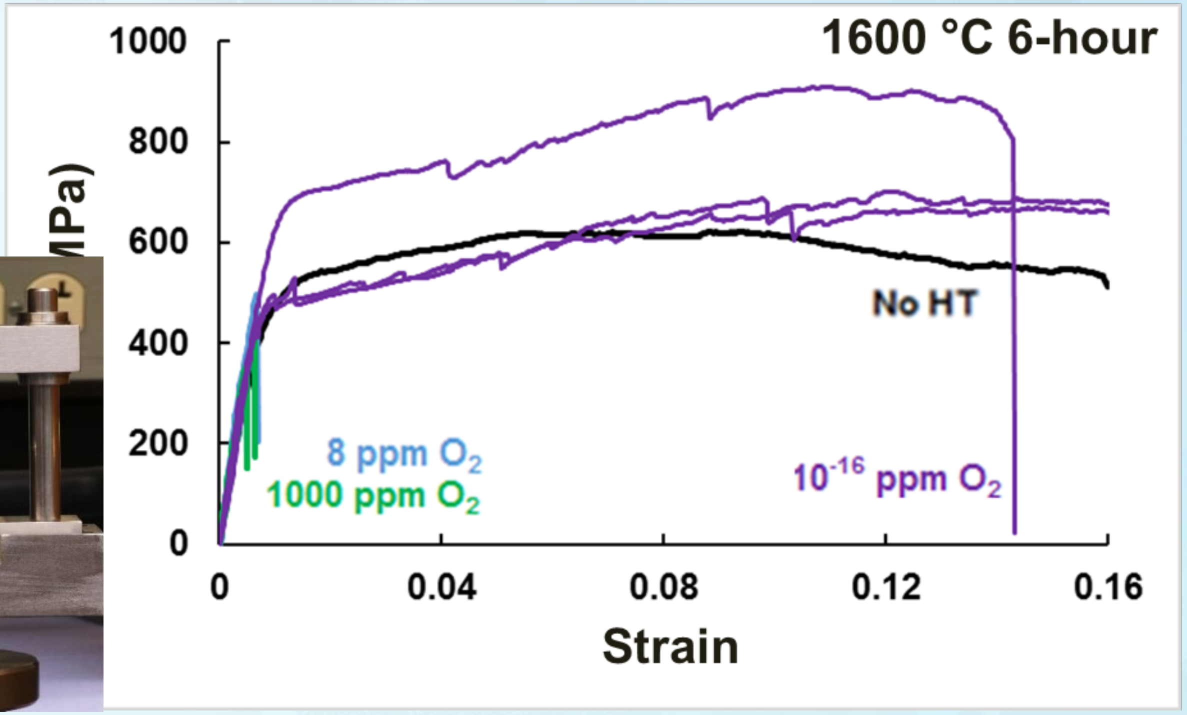
1. "Nuclear Thermocouple Drift models." S. Riley, R. Skifton, B.J. Jaques. *In Draft*.
2. "Methods for Temperature and Thermal Conductivity Measurements in Extreme Environments." K. Wada, S. Riley, B.J. Jaques, D. Estrada. *In Draft*.
3. "Influence of Microstructure and Phase Morphology on the Stability of High temperature Irradiation Resistant Thermocouples." S. Riley, K. Holloway, A. Bateman, R. Skifton, B.J. Jaques. Materials Today Communications. 2023. *In Press*.
4. "Development of drift models for high temperature nuclear thermocouples." S. Riley, R. Skifton, B.J. Jaques. Advanced Sensors and Instrumentation Newsletter. Issue 17. September, 2022

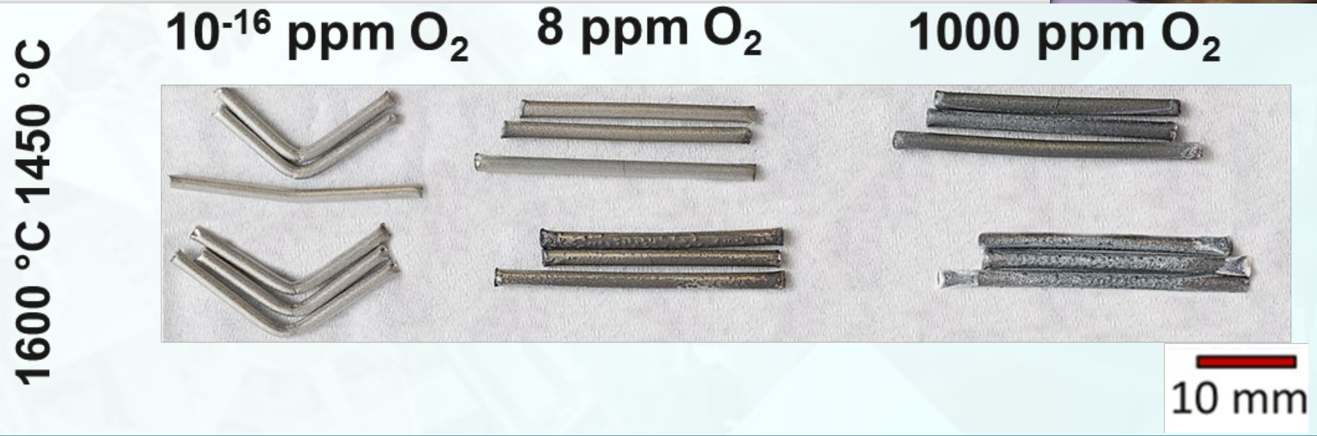
Results and Accomplishments

Effect of process temperature and PO_2 on HTIR-TC ductility via 3-point bending







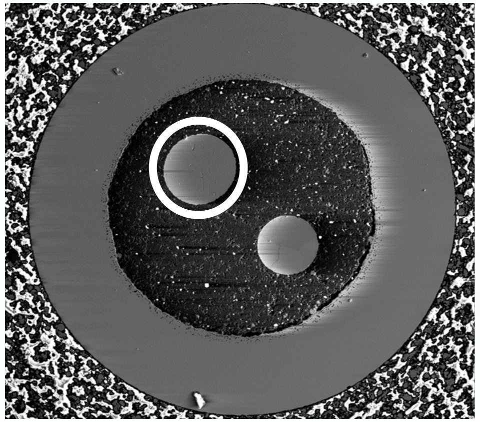


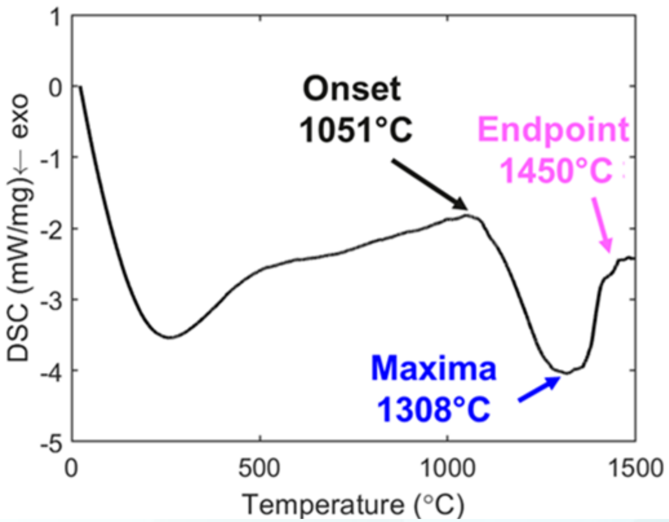
Finding: Heat treatment with $PO_2 \geq 8$ ppm resulted in Nb sheath embrittlement.

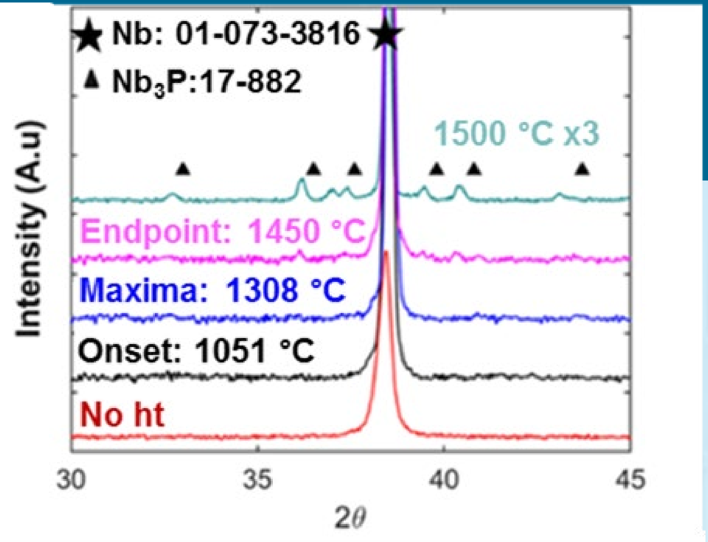
Solution: Perform stabilization heat treatment in either a sacrificial Nb foil to “getter” O_2 or very clean atmosphere to retain ductility

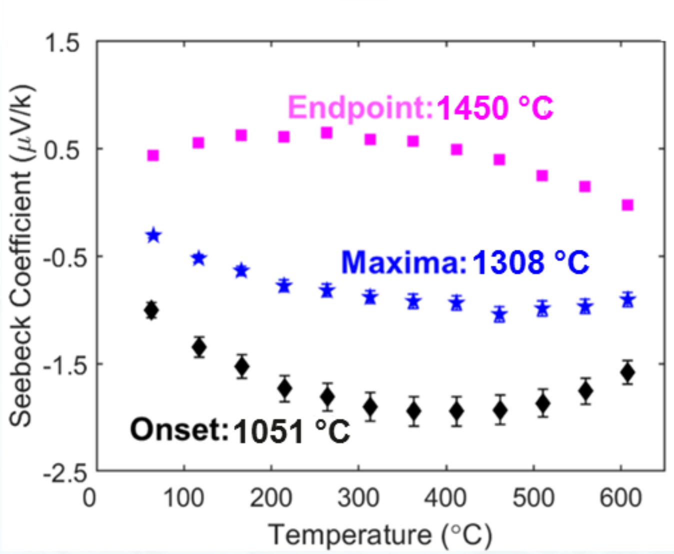
Results and Accomplishments

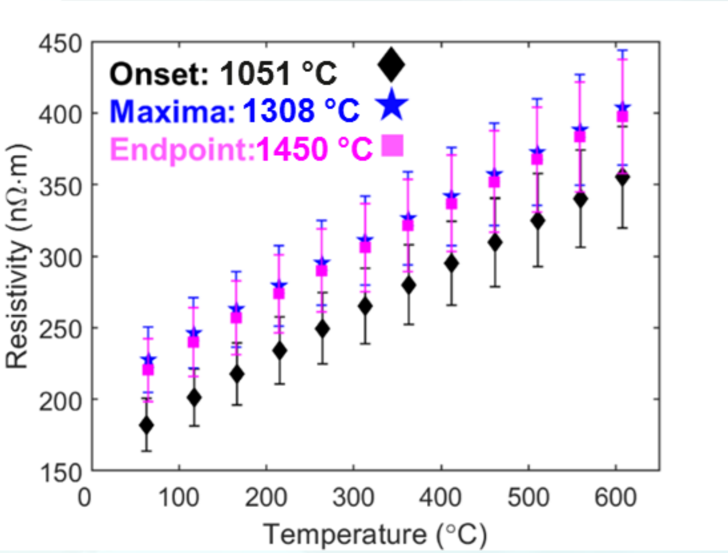
Characterization of Bare Nb-P Thermoelement

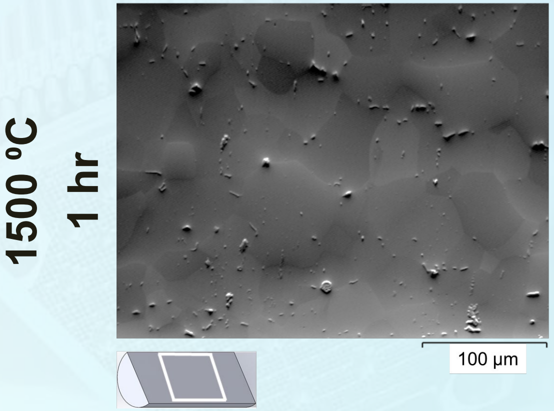


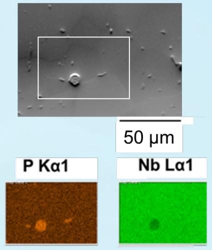










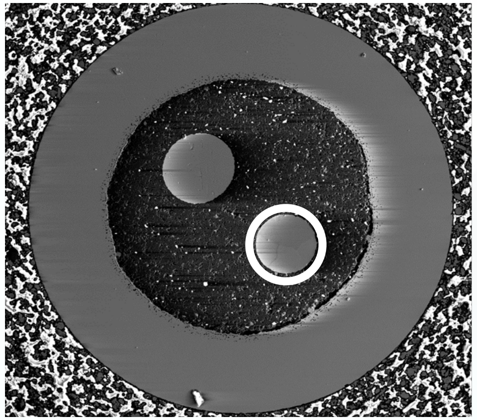


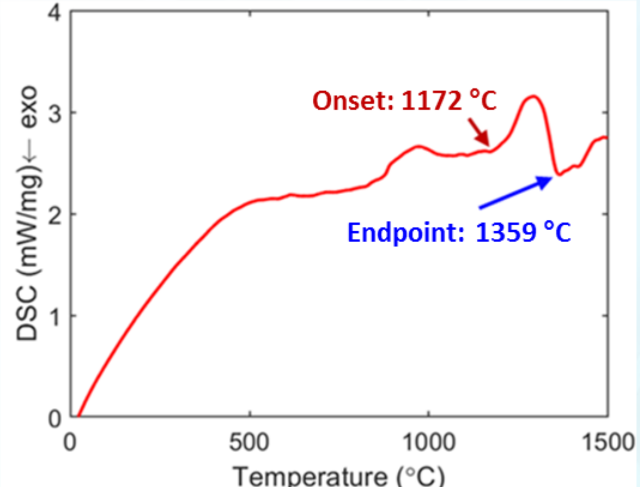
Findings: A non-reversible exothermic reaction occurs in the Nb-P thermoelement where Nb_3P precipitates form throughout and alters the Seebeck coefficient of the TC

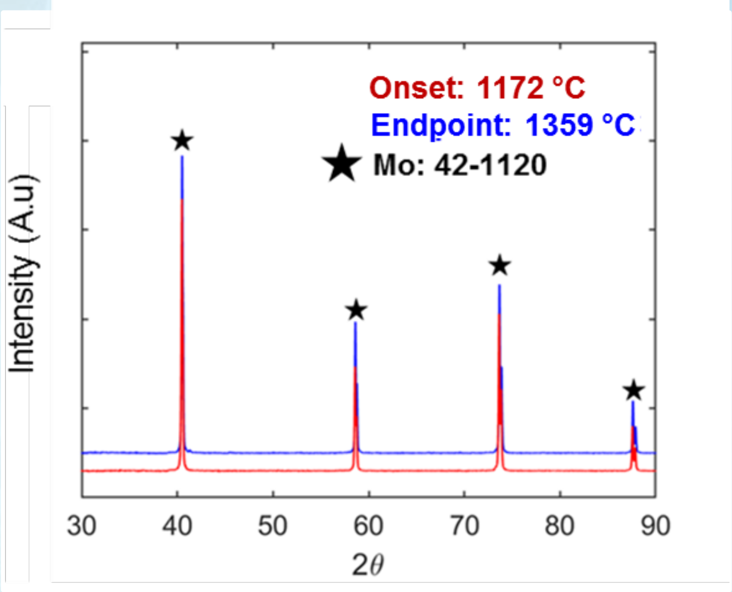
Impact: A stabilization heat treatment of >1450 °C is necessary to reduce Nb-P thermoelement drift during operation by stabilizing nucleation and growth.

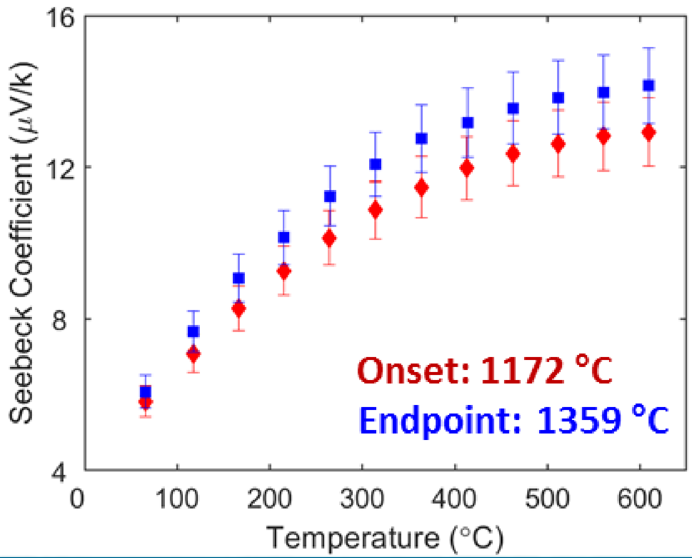
Results and Accomplishments

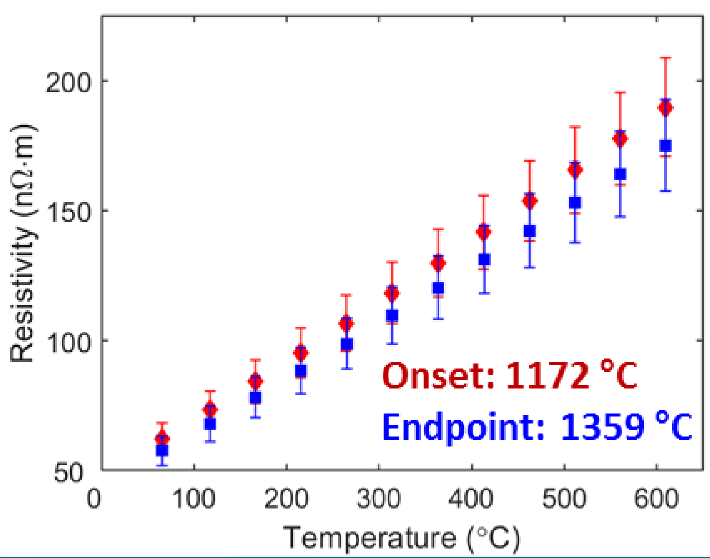
Characterization of Bare Mo-LaO Thermoelement









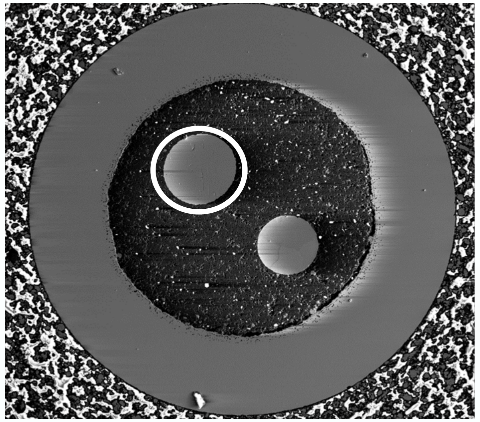


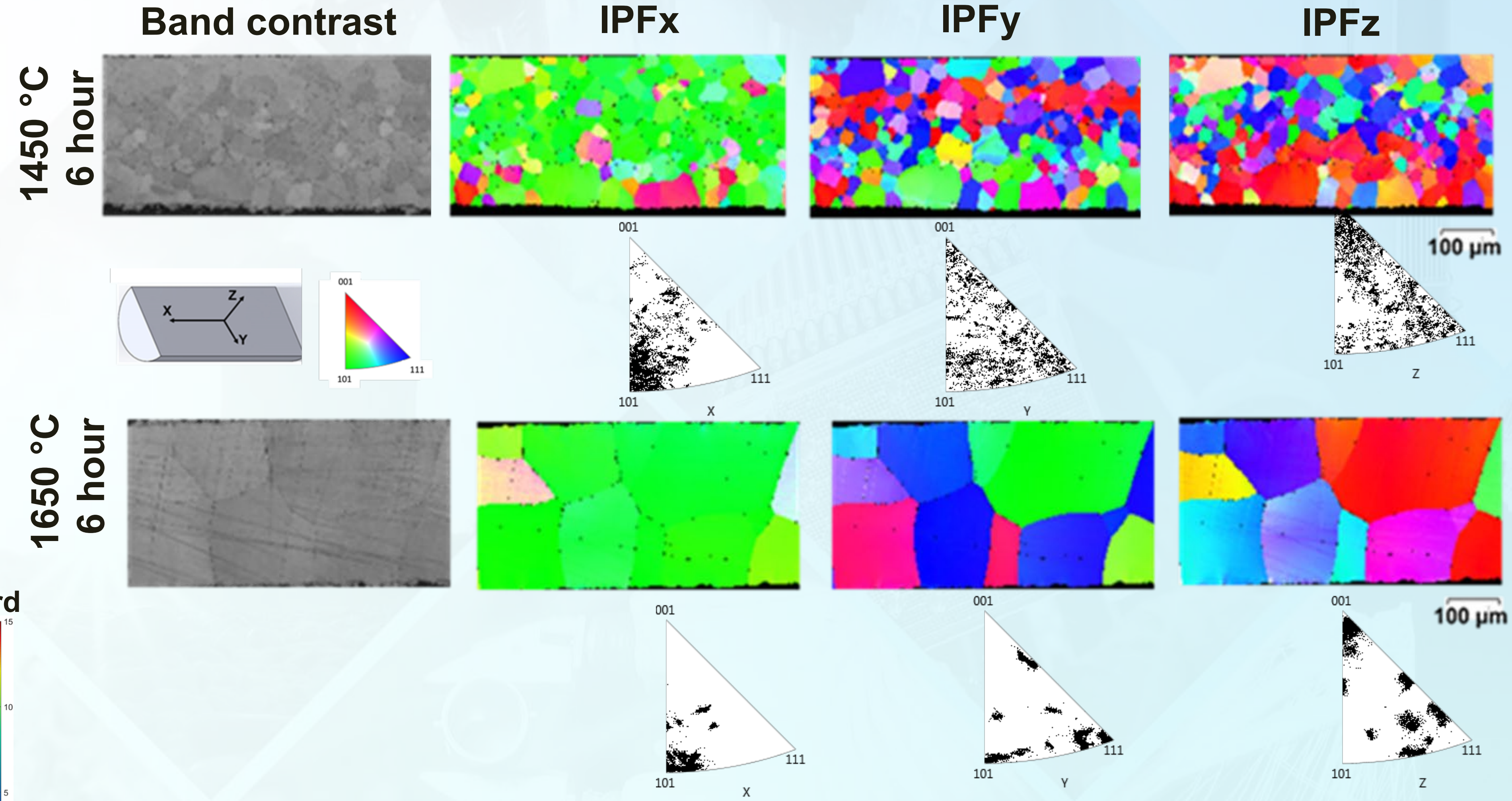
Findings: A reversible endothermic reaction occurs in the Mo-LaO thermoelement during heat treatment; however, the Seebeck, electrical resistivity, and chemical phases of the Mo thermoelement remain stable throughout the heat treatment.

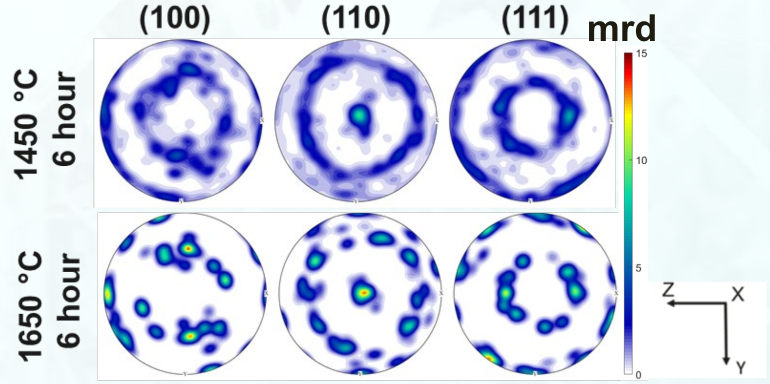
Impact: The Mo thermoelement has minimal contribution to thermal drift.

Results and Accomplishments

Mechanistic investigation of the Nb-P thermoelement drift via grain morphology evolution.



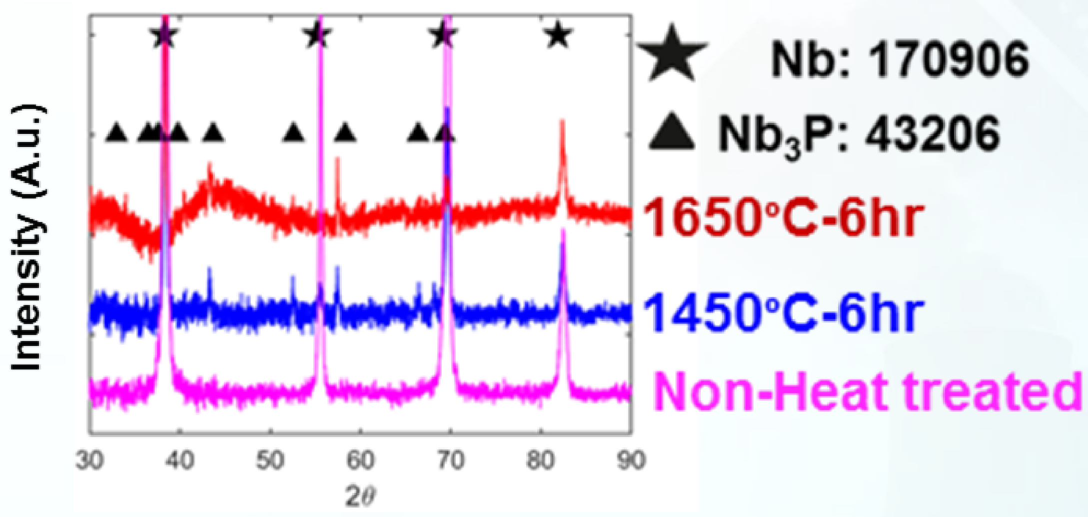


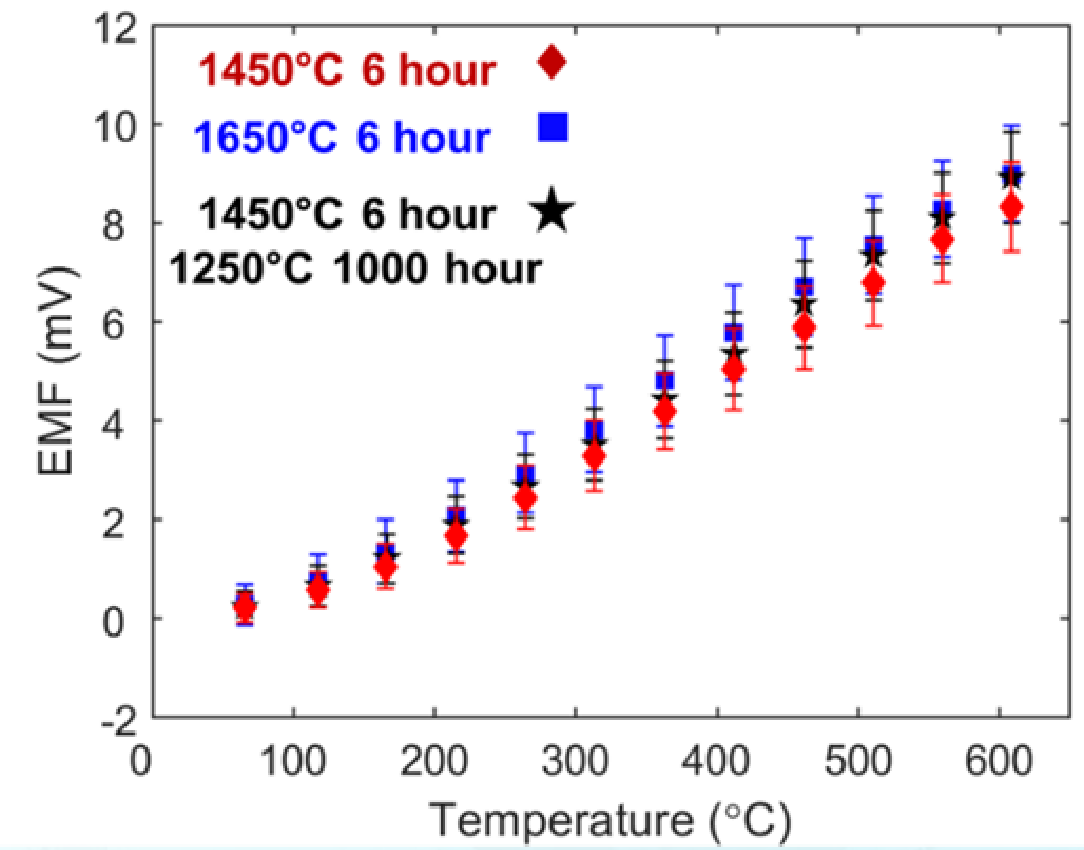


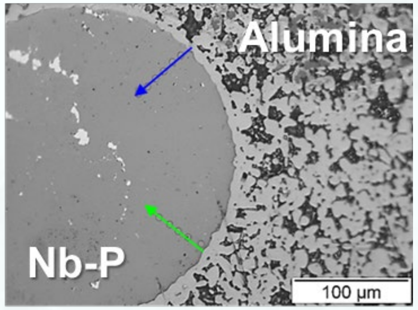
The Nb-P thermoelements do not recrystallize when heat treated at either 1450 or 1650 °C but rather, fiber texture is retained and exacerbated through grain growth.

Results and Accomplishments

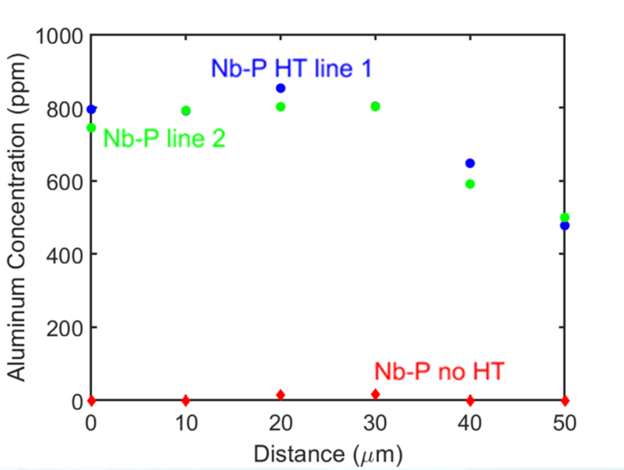
Chemical Stability of Nb-P Thermoelement







1600 ºC - 24hr - UHV



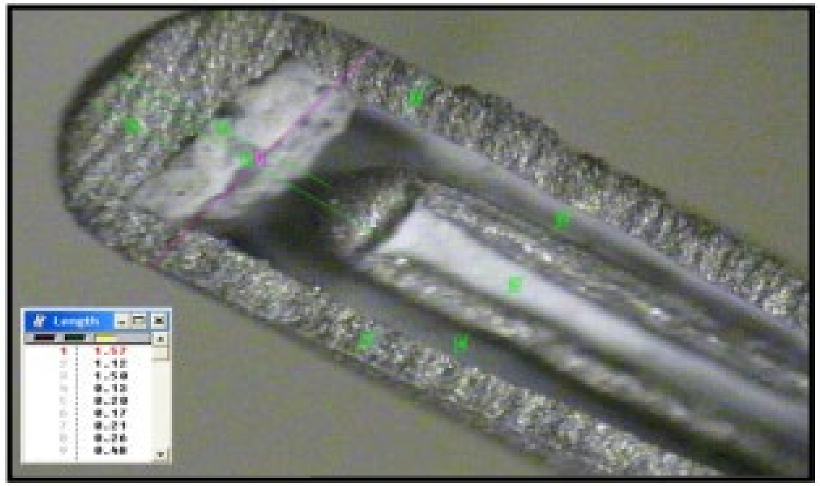
Findings: Alumina reacts with Nb-P where it dissociates and diffuses through the Nb-P thermoelement; however, once it is formed, it appears to be stable

Impact: Stabilization heat treatment induces mass diffusion and therefore reduces driving force for continued diffusion (contributing to thermal drift) especially when operated at lower than heat treatment temperatures.

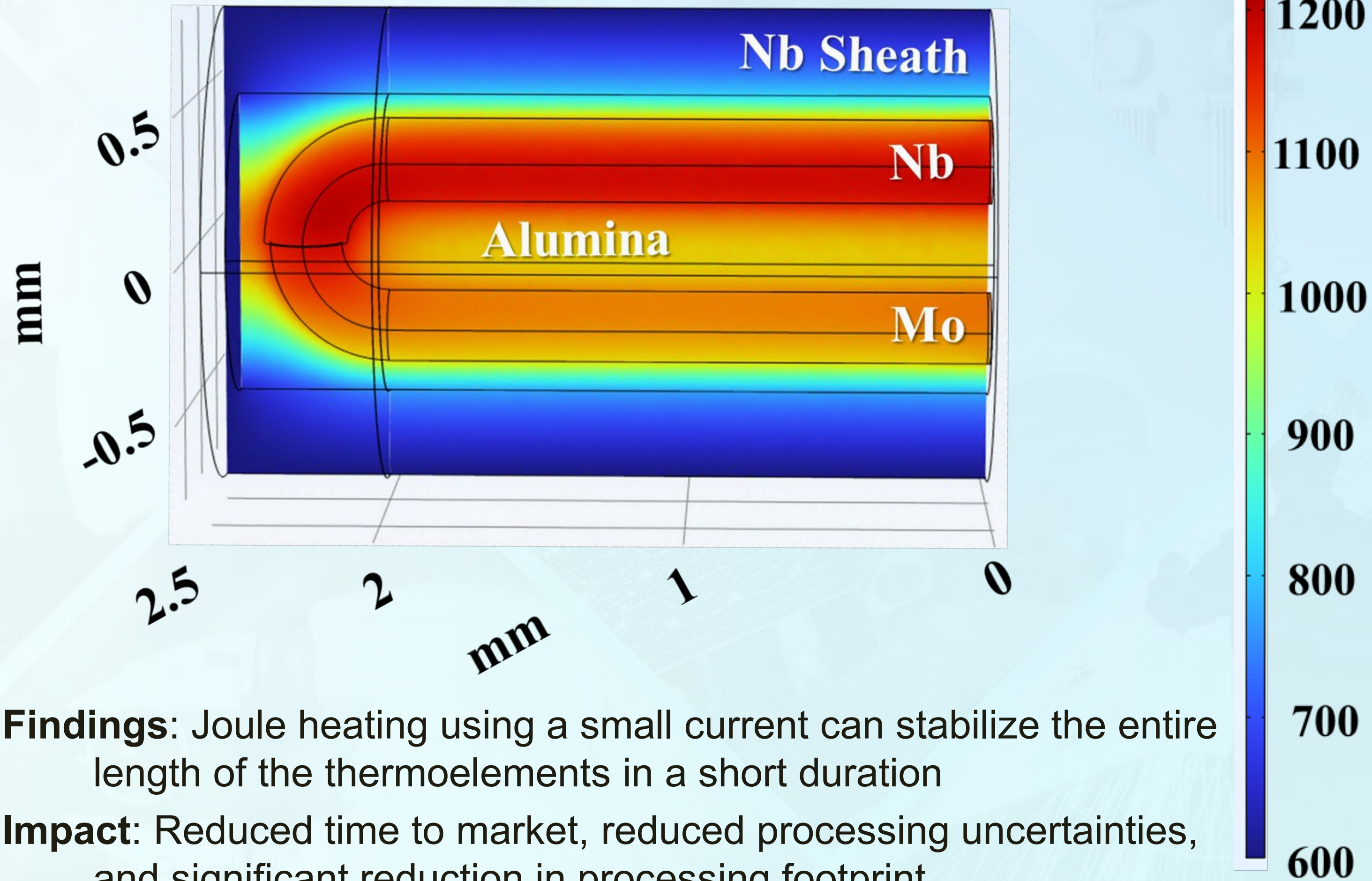
Results and Accomplishments

Modeling of Alternative Heat Treatment Methods of HTIR-TCs

Thermocouple Hot Junction[1]



Joule heating model of HTIR-TC hot junction cross-section in COMSOL[2]



HTIR-TC Component	Average Temperature (° C)
Mo	1068
Nb	1139
Mo Hot Junction	1113
Nb Hot Junction	1149

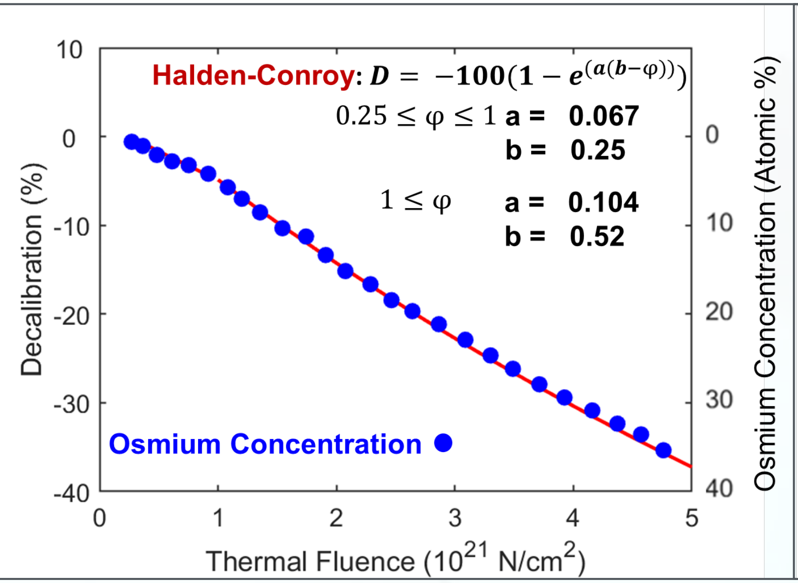
Findings: Joule heating using a small current can stabilize the entire length of the thermoelements in a short duration

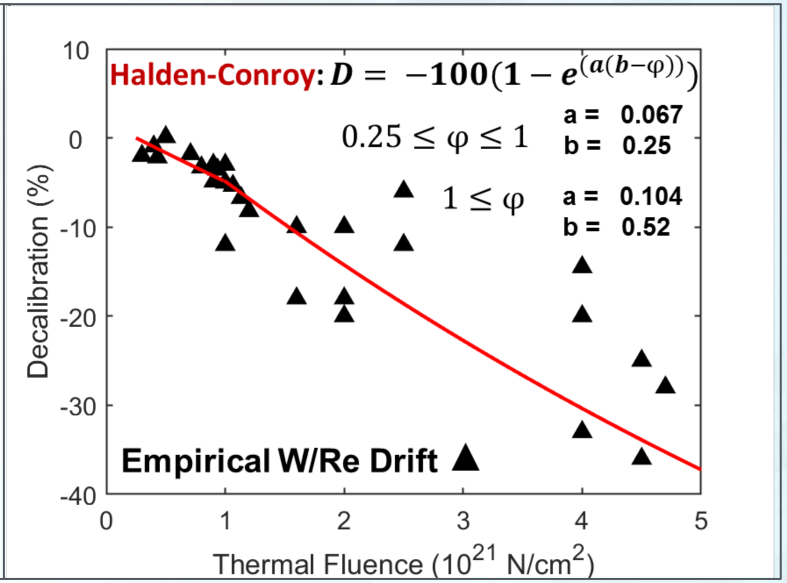
Impact: Reduced time to market, reduced processing uncertainties, and significant reduction in processing footprint

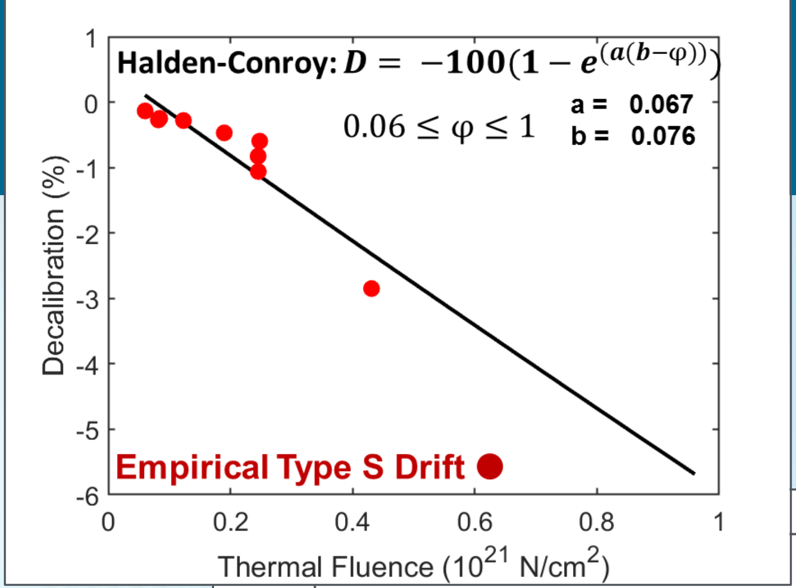
1. M. Scervini et. al. Low drift type K and N mineral inslated thermocouples for high temperature applications. Department of Materials Science and Metallurgy, University of Cambridge.
2. Multiphysics, C., 1998. Introduction to COMSOL multiphysics extregistered. COMSOL Multiphysics, Burlington, MA, accessed Feb, 9, p.

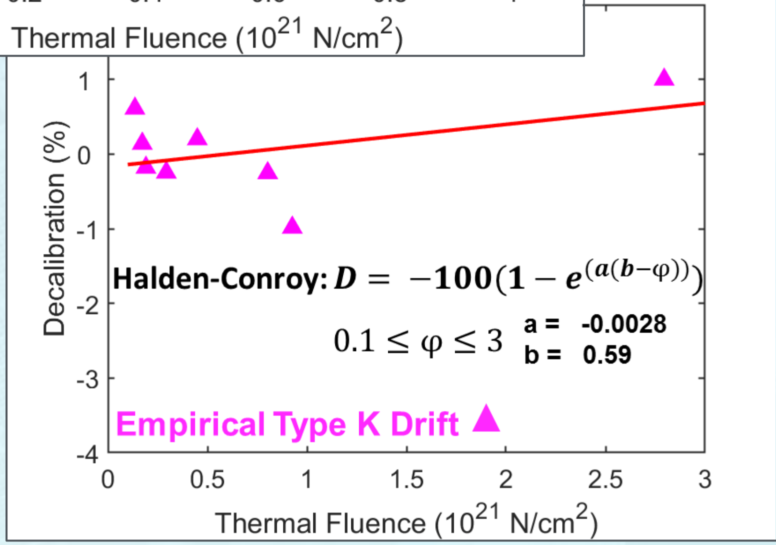
Results and Accomplishments

Empirical Thermocouple Drift Model









Where φ is the thermal neutron fluence (in units of 10^{21} N/cm^2), and a/b are fitting coefficients

Thermocouple Type	Halden-Conroy Decalibration Equation	Thermal Fluence Range (10^{21} N/cm^2)	Decalibration Coefficients *a*	Decalibration Coefficients *b*	References
C	$D = -100(1 - e^{(a(b-\varphi))})$	$0.25 \le \varphi < 1$	0.067	0.25	[3, 4, 6, 7, 8]
		$1 \le \varphi$	0.104	0.52	
S	$D = -100(1 - e^{(a(b-\varphi))})$	$0.06 \le \varphi \le 1$	0.067	0.076	[3, 6, 10]
K	$D = -100(1 - e^{(a(b-\varphi))})$	$0.1 \le \varphi \le 3$	-0.0028	0.59	[3, 6, 7]

Findings: The Halden-Conroy empirical drift model (transmutation is largest contributor to drift) describes the empirical behavior of S- and K-type TCs.

Impact: Can be applied to other TCs (e.g. HTIR-TCs) to predict the rate of decalibration due to irradiation. This model can be coupled with thermal drift behavior to accurately predict high temperature TC behavior in advanced reactor environments.

Concluding Remarks

HTIR-TCs marry the high temperature capability with the stability necessary for nuclear applications.

- Stabilization heat treatments are necessary to reduce (not quite eliminate) thermal drift
- The ductility of the HTIR-TC build can be retained by:
  - Precisely controlled atmospheres during heat treatment (i.e. low PO_2)
  - Joule heating resulting in rapid heat treatments and low sheath surface temperatures
- Nb-P thermoelement is the primary culprit in HTIR-TC drift due to:
  - Precipitation of Nb_3P
  - Nb – Alumina interaction region that results in dissociation and diffusion of aluminum throughout the thermoelement.
- However, these phenomena can be stabilized by a heat treatment >100 °C above the max service temperature

- This work was/will be disseminated through:
  - 2 journal publications
  - 2 manuscripts that are in draft
  - 4 conference presentations
  - 1 upcoming conference presentation
  - 1 ASI newsletter
  - Scott Riley's PhD Dissertation (TBD)

Future Work

- Refine COMSOL models using geometries obtained from XRCT scans of typical builds
- Create a physical basis for the decalibration fitting coefficients determined for type S and K thermocouples (e.g. Do they have a strong correlation to transmutation product conc.)
  - Apply empirical drift model to HTIR-TC designs and observed behavior
- Optimize the stabilization heat treatment via Joule heating through mechanical and electrical properties, chemical stability, and microstructural evolution.

Scott Riley
Graduate Research Assistant at BSU and INL Intern

Thank You

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Fellow | Center for Advanced Energy Studies
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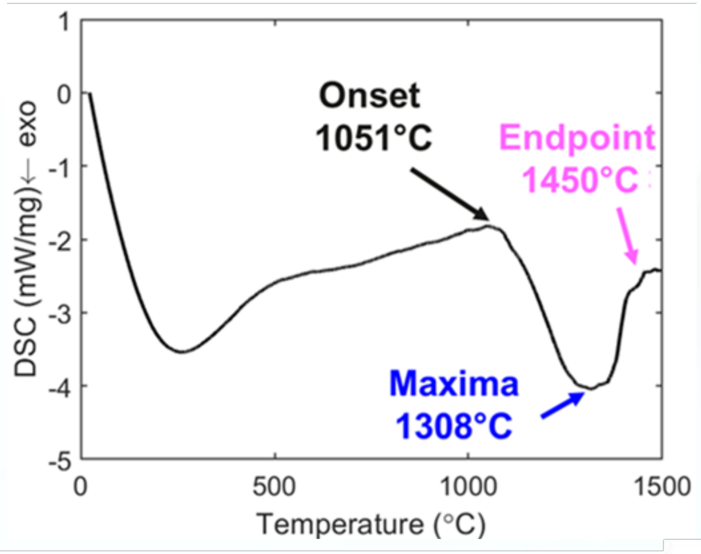
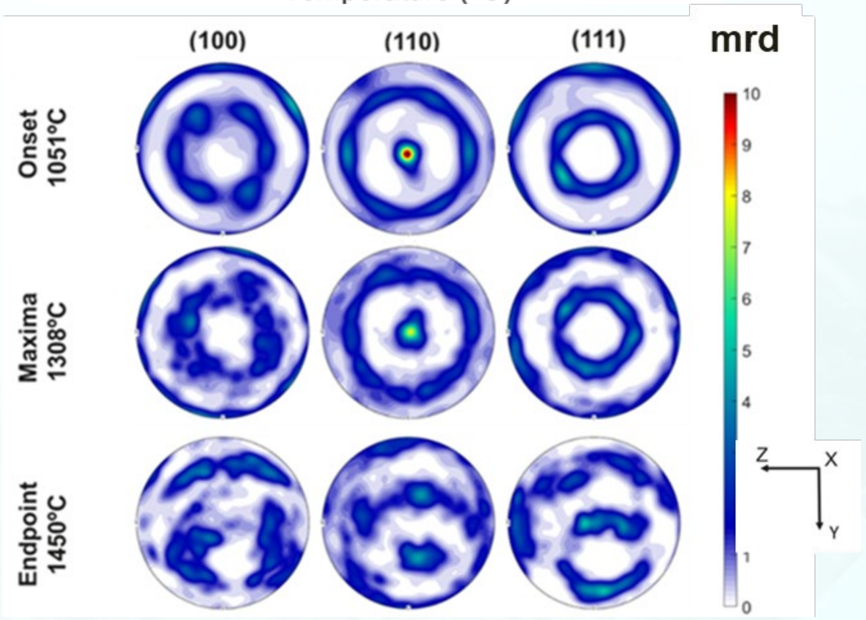
AML
Advanced Materials Laboratory

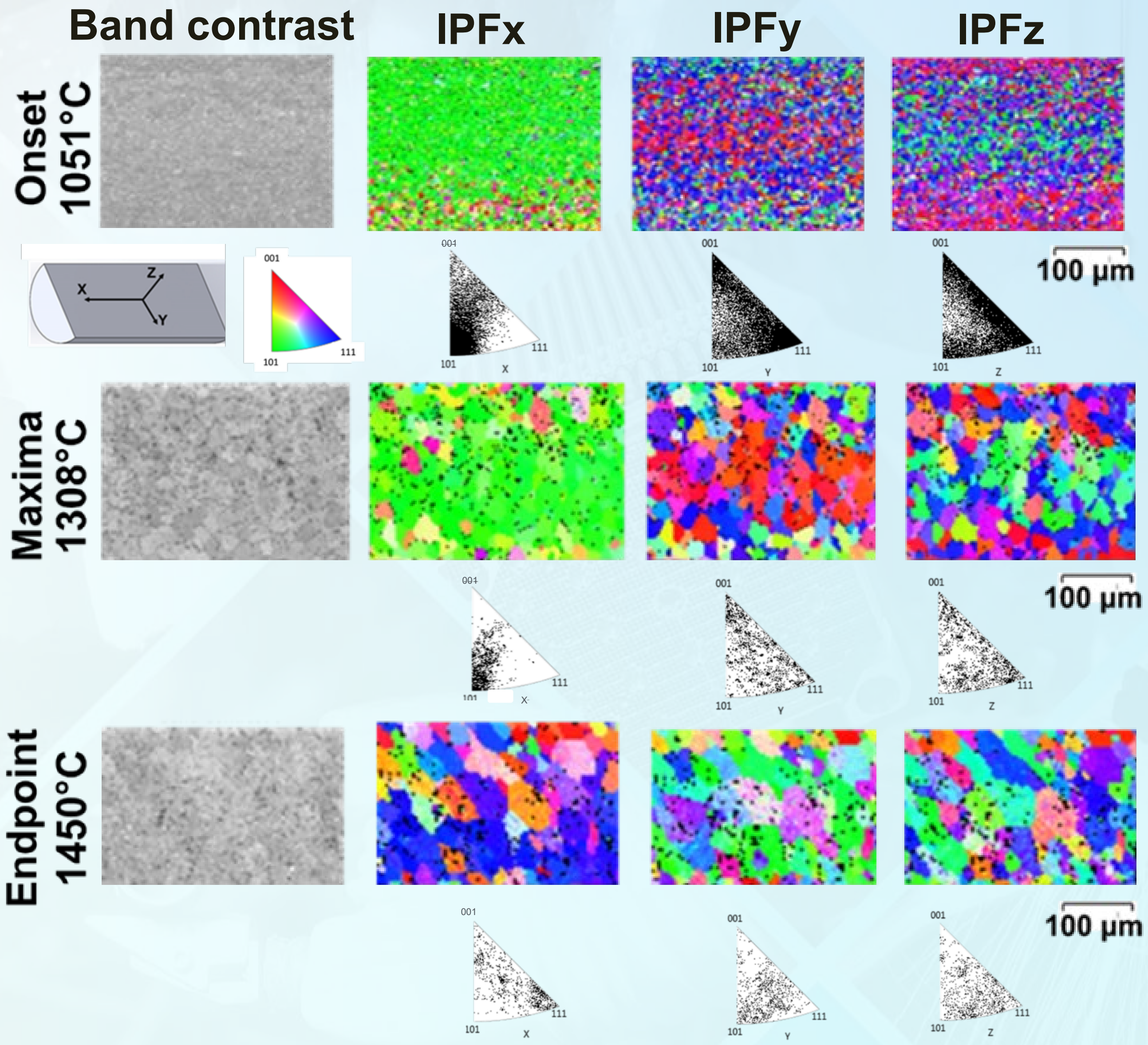
Empirical Drift Model References

[2] B. Conroy, The Calculation of perturbed fluece in HFO/Y2O3 end pellets for thermocouple decalibration purposes, HWR-181 (1986).
[3] C. Patterson et al. Thermocouple decalibration-an interim recommendation, General Electric Report NEDO-30673 (1984).
[4] R. Pratt, An investigation of the effects of transmutation on the thernio-electric stability of W5%Re-W26%Re thermocouples, AERE-M 2081 (1968).
[5] C. Vitanza, T.E. Stien, ASSESSMENT OF FUEL THERMOCOUPLE DECALIBRATION DURING IN-PILE SERVICE, Journal of Nuclear Materials 139(1) (1986) 11-18.
[6] R. Van Nieuwenhove, L. Vermeeren, Irradiation Effects on Temperature Sensors For ITER Application, Review of Scientific Instruments 75(1) (2004) 75-83.
[7] N.L. Sandefur, J.S. Steibel, R.J. Grenda, Emf Drift of Chromel/Alumel and W3%/W25%Re Thermocouples Measured in Pile to High Neutron Exposures, American Nuclear Society Annual Meeting (Gulf-GA-AL2501) (1973) 1-19.
[8] J.Heckelman, R. Koza, Measured drift of irradiated and unirradiated W-3%Re/W-25%Re thermocoupless at a nominal 2000K, Lewis Research Center Report T3-13 (1971).
[9] W.Browning, C. Miller, Calculated radiation induced changes in thermocouple composition, Proceedings of the 4th Symposium on Temperature: Its Measurement and Control in Science and Industry 2 (1961) 271.
[10] M.J. Kelly, W.W. Johnston, C.D. Baumann, The Effects of Nuclear Radiation on Thermocouples, Temperature Its Measurement and Control in Science and Industry 3 (1962) 265-269.

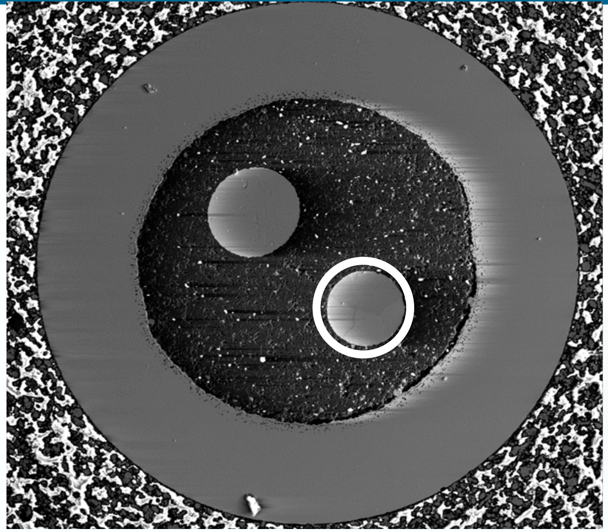
Characterization of Bare Nb-P Thermoelement

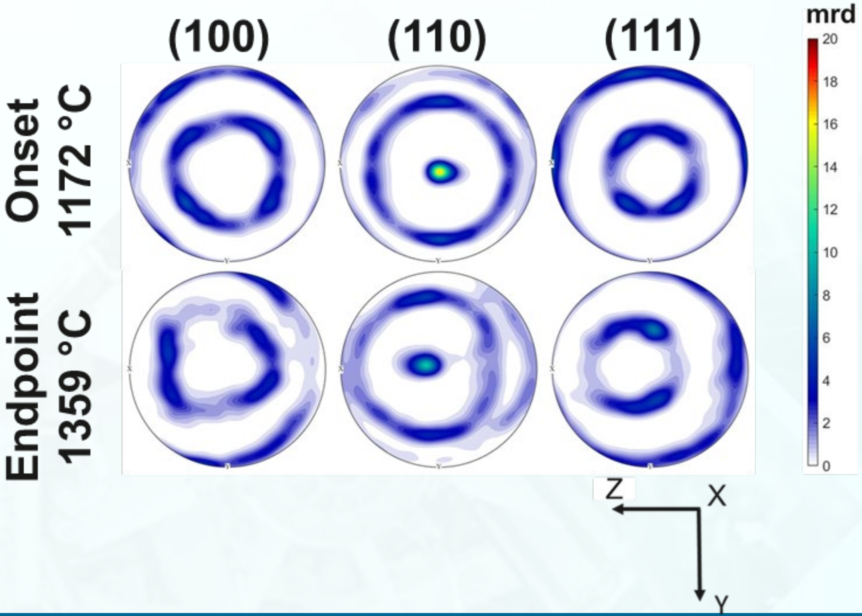


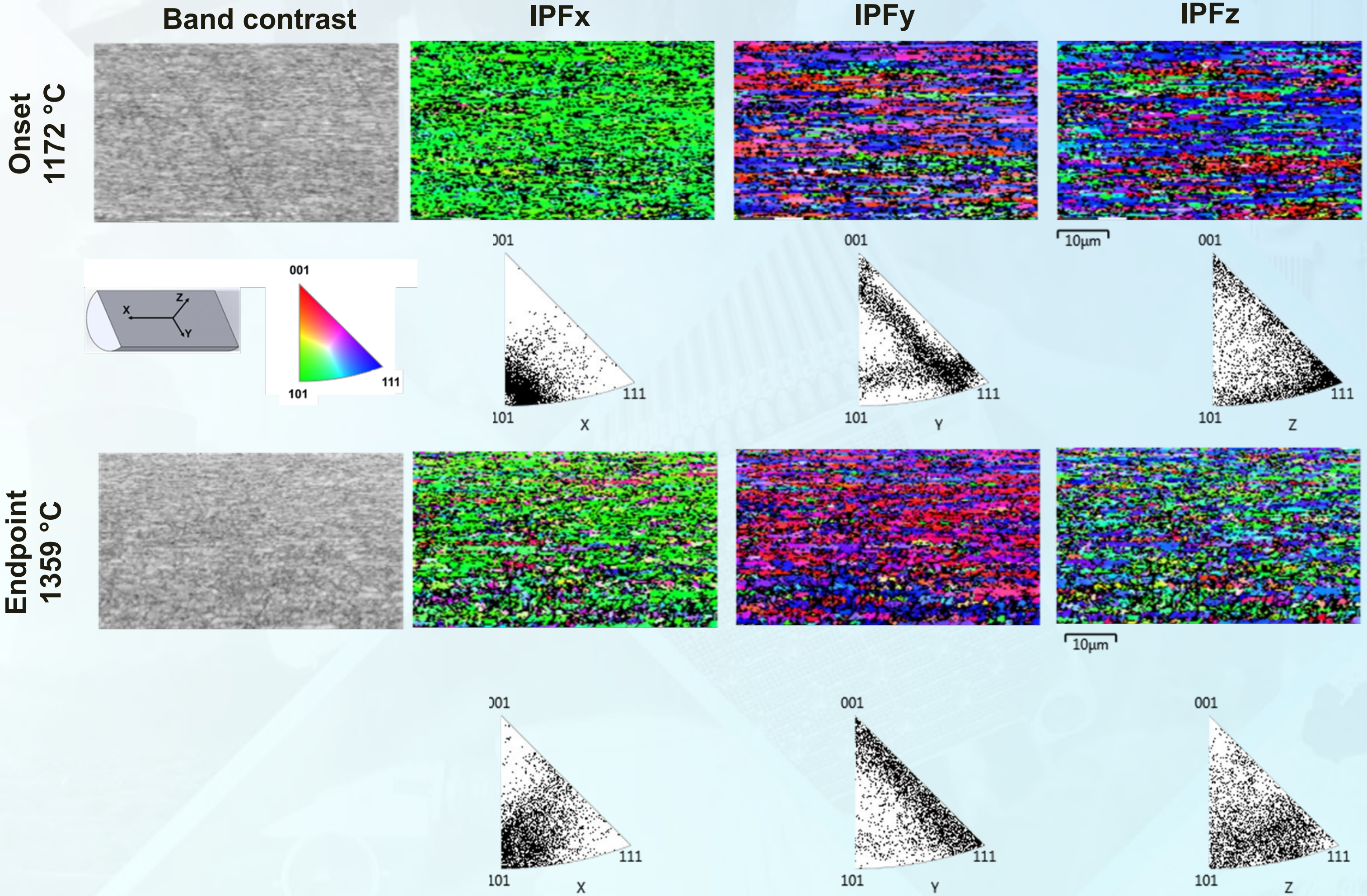




Characterization of Bare Mo-LaO Thermoelement

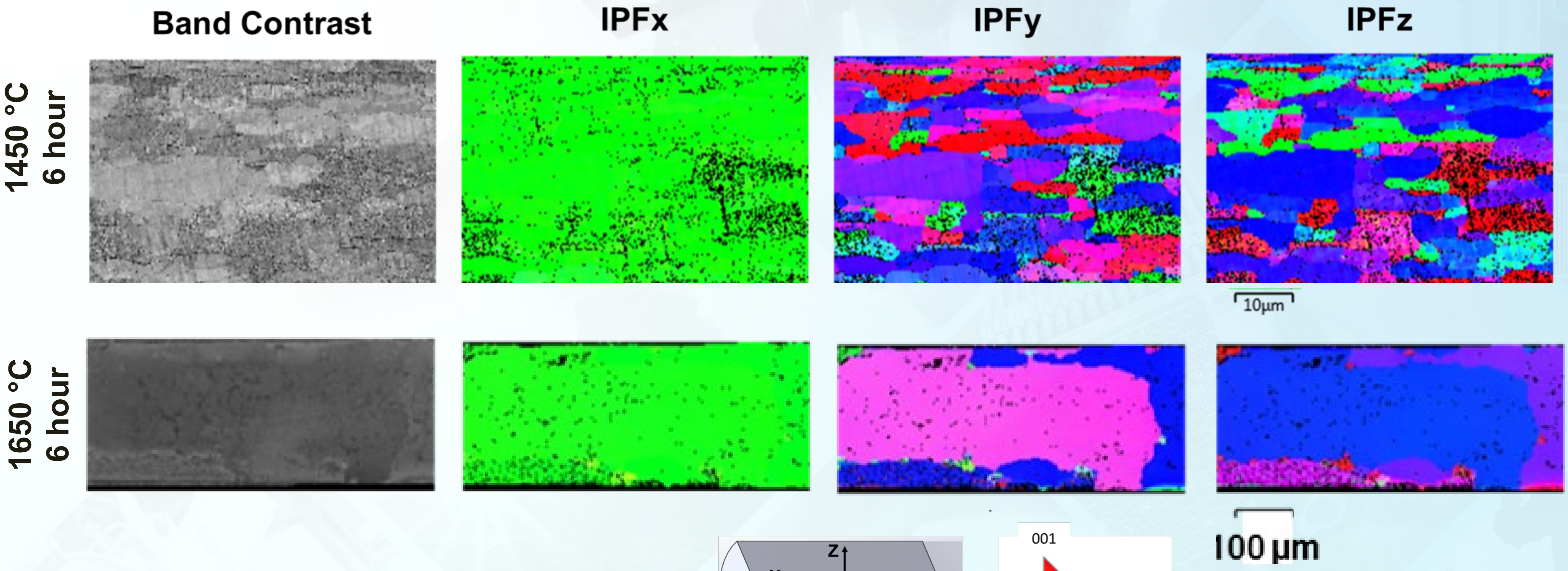


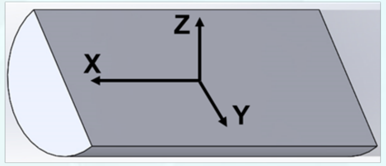


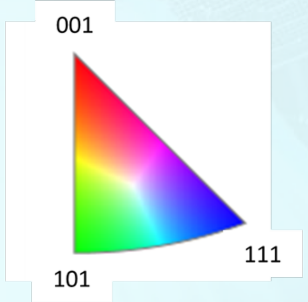


- The (101)//x preferential orientation is largely retained across the endothermic transformation.

Stability of Mo-LaO Thermoelement







- Heat treatment at either 1450 or 1650 °C for 6 hours did not result in recrystallization of the Mo-LaO thermoelement.