U.S. Nuclear Regulatory Commission's
35th Annual Regulatory Information Conference

Project:
Advanced characterization of ATF cladding for understanding their degradation under short-time temperature excursions and implications in dry storage

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➢Nanomaterials Characterization Core Facility at VCU

➢Ge Global Research (Andrew Hoffman), Rajnikant Umretiya

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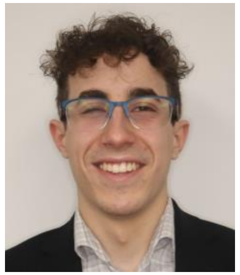

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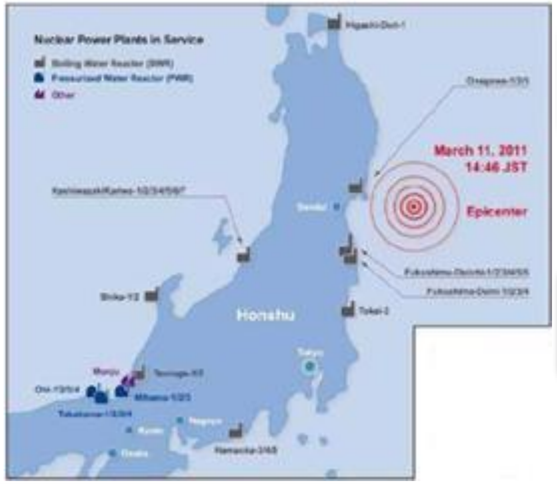
Moving on to the topic of today's talk…

- Fukushima nuclear disaster

(AP Photo/Yomiuri Shimbun, Masamine Kawaguchi)

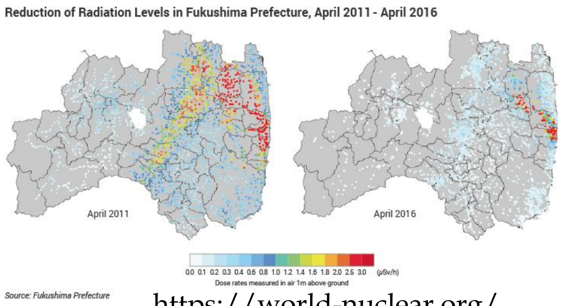


Earthquake

+

Tsunami

radioactive release



https://world-nuclear.org/

Zircaloy

$Zr + 2\ H_2O \rightarrow ZrO_2 + 2\ H_2$

Exothermic reaction and release of hydrogen

Hydrogen explosion

How to improve the safety and reconstruct the public's confidence in nuclear power?

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The Fukushima Daiichi Accident Report by the Director General (2015)

Accident Tolerant Fuel Technologies

ATF Claddings

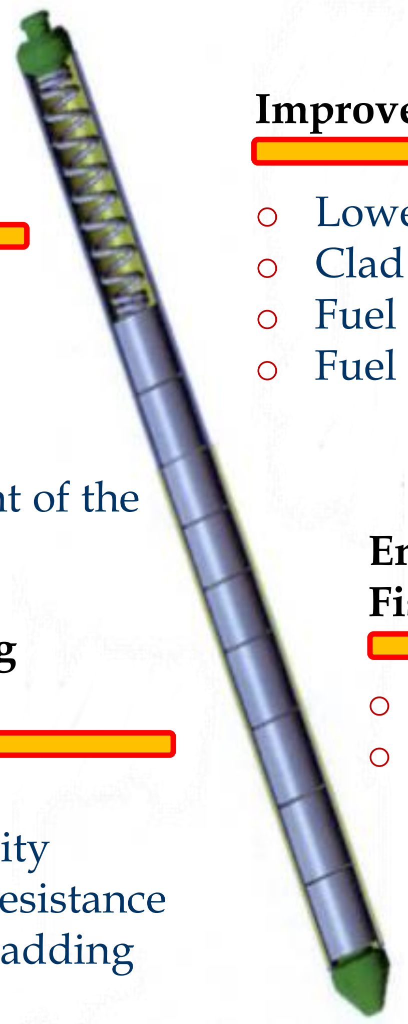
Surface modification	Replacement
Concept 1	Concept 2
Zr alloys, Surface coating	Alloys
FeCrAl	APMT
Cr	C26M
Mo	310SS
SiC	

Improved Clad Reaction Kinetics with Steam

- Heat of Oxidation
- Oxidation rate
- Hydrogen bubble and explosion
- Hydrogen embrittlement of the clad

Improved Cladding Properties

- Clad fracture
- Geometric stability
- Thermal shock resistance
- Melting of the cladding



Improved Fuel Properties

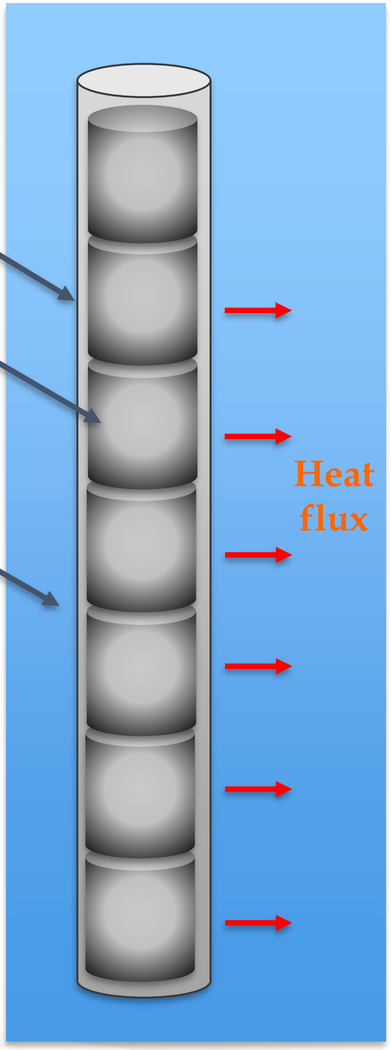
- Lower operating temperatures
- Clad internal oxidation
- Fuel relocation
- Fuel melting

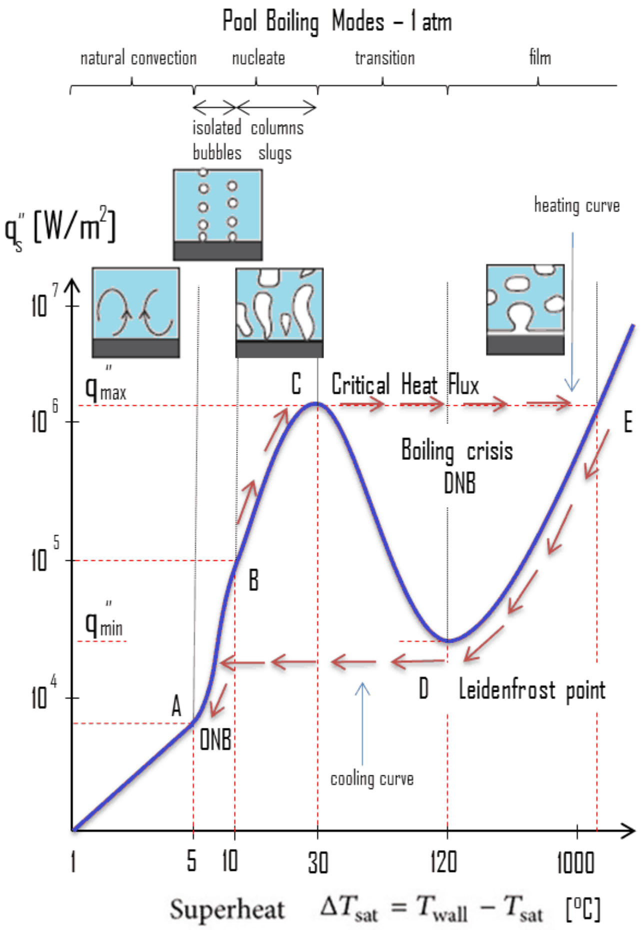
Enhanced Retention of Fission Products

- Gaseous fission products
- Solid/liquid fission products

Areva's Enhanced Accident Tolerant Fuel Program (2017)

How do these cladding concepts evolve at high temperatures?

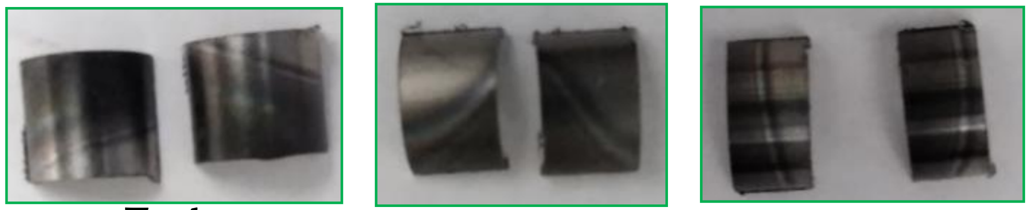




The following reactor parameters affect safety margins:

- Reactor power
- Reactor coolant flow rate
- Reactor coolant inlet temperature
- Reactor coolant pressure

How does the material evolves at high temperatures and how quick?



Surface characterization before and after CHF testing

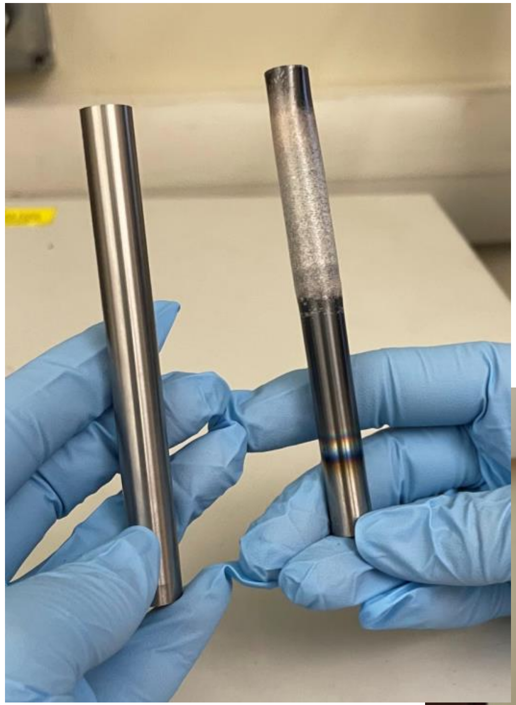
Umretiya, R.V., Elward, B., Lee, D., Anderson, M., Rebak, R.B. and Rojas, J.V., 2020. J. Nuc. Mat., 541, p.152420.

Project Goals

The main goal of this project is to investigate the oxidation and degradation of accident-tolerant fuel (ATF) claddings, both Cr-coated Zircaloy and FeCrAl alloys, under short time temperature excursions and quenching performance that will reveal their behavior and evolution during accident scenarios.

This project also investigates the mechanical properties of these materials relevant to dry storage conditions. Implementing (ATF) cladding in light water reactors (LWRs) and advanced reactor designs aims to improve fuel reliability and safety during design-basis and beyond-design-basis accident scenarios.

Implementation of non-destructive examination techniques for quality control of coated Zircaloy.



Zr-4 subjected to rapid heating and cooled in air or water

Research Objectives

Aim 1: ATF cladding selection, sample manufacturing, and characterization:

- Use various advanced materials characterization techniques to study surface characteristics microstructure, materials surface chemistry, mechanical properties

Aim 2: Short-time temperature excursions and quenching:

- We will investigate cylindrical specimens of both pristine and aged specimens
- The aging process: samples exposed to BWR and PWR using a hydrothermal autoclave
- The specimens will be subjected to high temperature heating profiles to peak temperatures ~1400 C

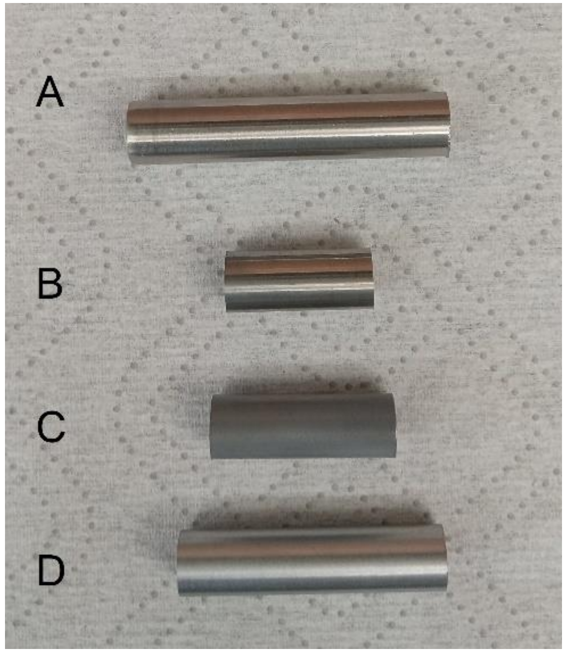
Aim 3: Simulated dry-storage conditions and ductility studies:

- The simulated dry storage conditions for the specimens: Interim Staff Guidance 11, Rev. “Spent Fuel Project Office

Aim 4-5: Development of X-ray fluorescence spectroscopy ATF cladding non-destructive examination:

- Design and construction of XRF setup for continuous analysis of cylindrical specimens The versatility of X-ray fluorescence spectroscopy will allow for simultaneous elemental composition and coating thickness analysis

Materials Selection and characterization



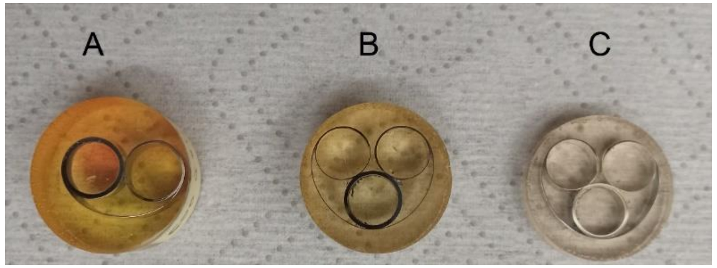
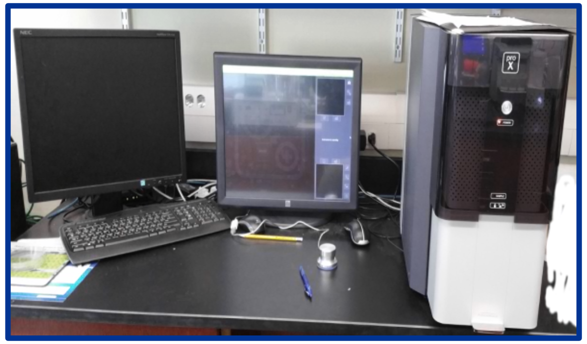


Figure 1. ATF cladding samples selected for materials characterization. In the photograph, A) FeCrAl alloy C26M, B) Zircaloy-4, C) Cr-coated Zr-4 prepared by PVD, D) Cr-coated Zr-4 prepared by Cold-spray



Phenom ProX SEM

- Scanning Electron Microscope (SEM) used to image sample surfaces
- Equipped with energy dispersive spectroscopy (EDS)

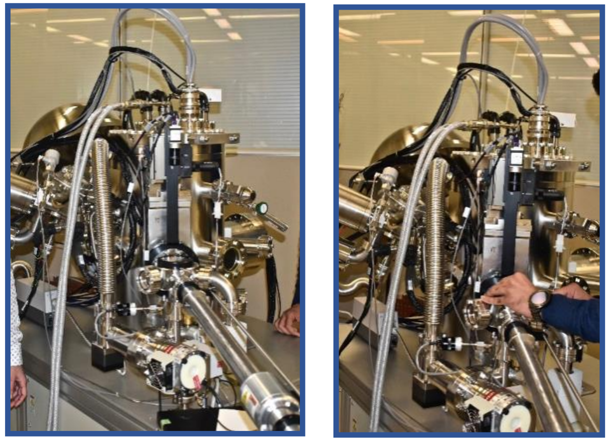
JEOL JEM-F200 TEM

- Transmission electron microscope for nanoscale analysis

Sample mounted on a powder specimen holder filled with **polyvinylpyrrolidone** (PVP) powder for **XRD** analysis

PANalytical X'Pert Pro Diffractometer

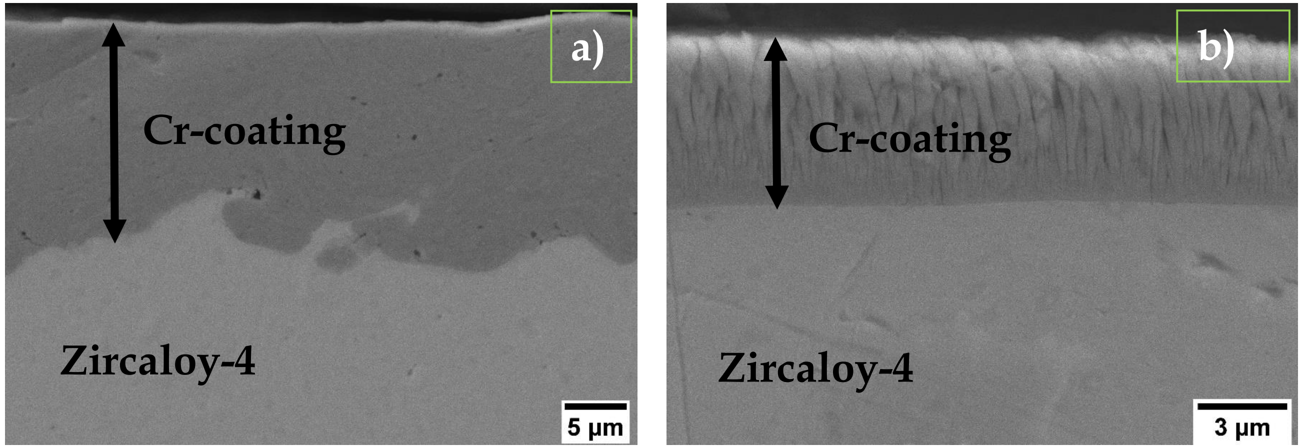
To obtain crystalline structure of materials



PHI VersaProbe III X-ray Photoelectron Spectrometer

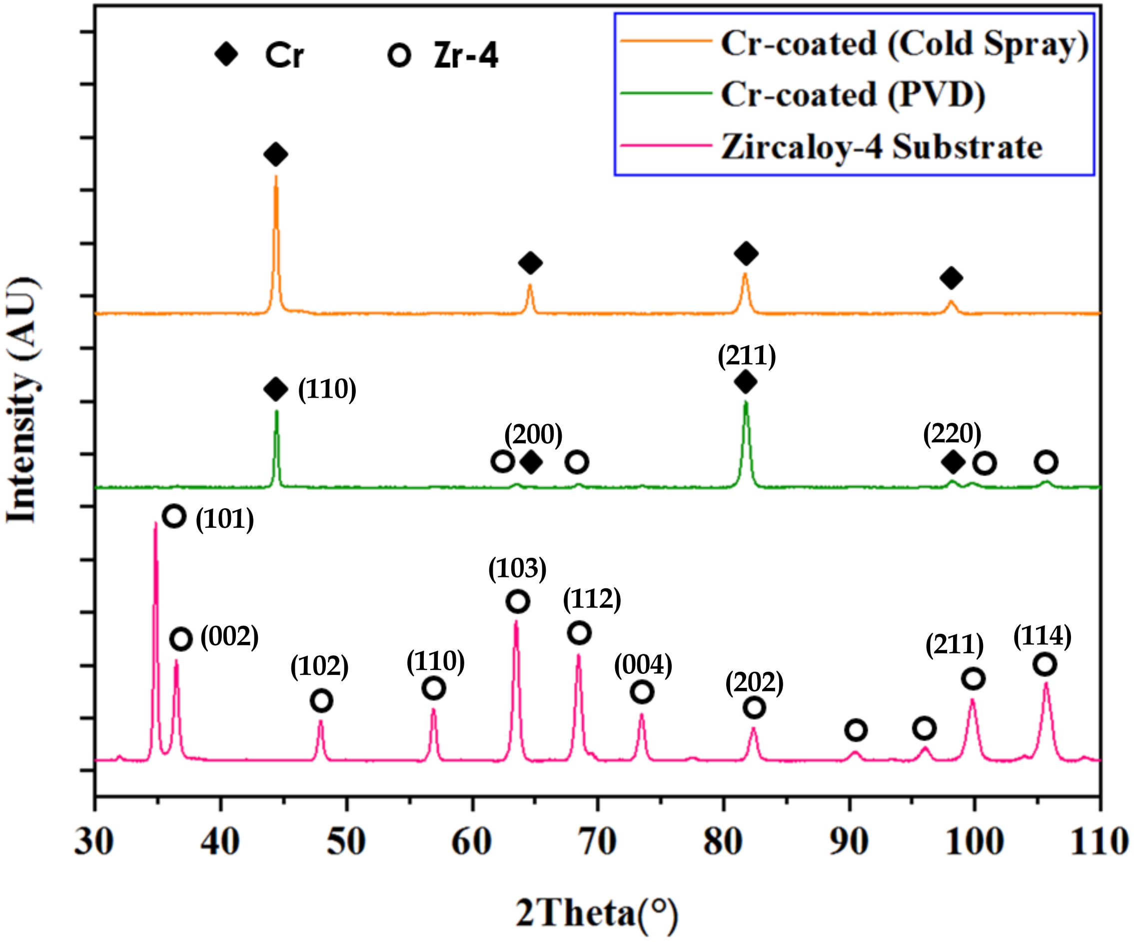
To study chemistry of sample surface

Coating deposition: Cr-Zr4



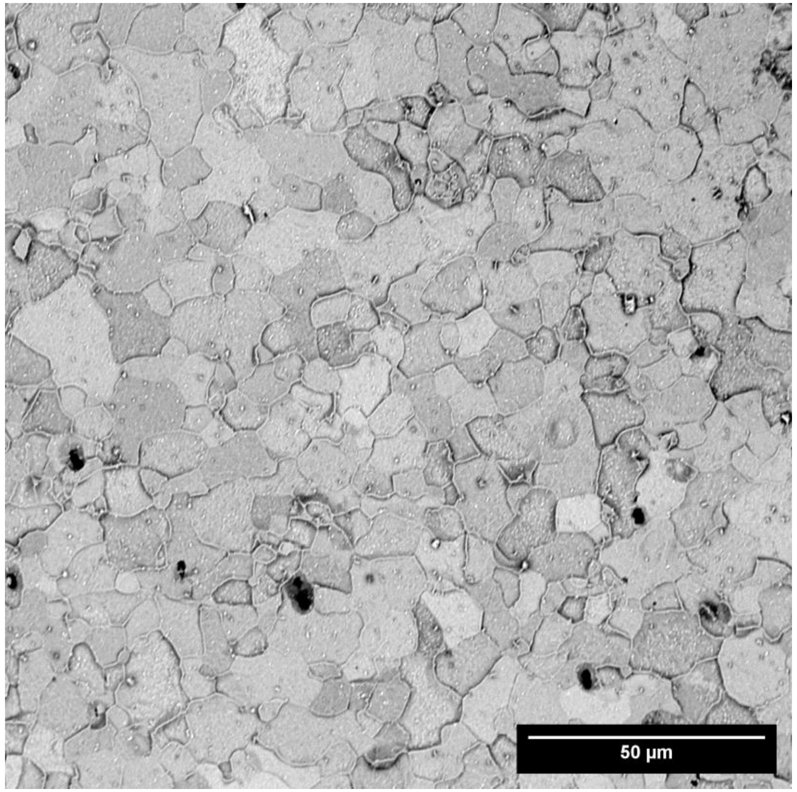
Cross sectional SEM view for Cr-coated Ziraloy-4: a) AR-Zr4-Cr-CS and b) AR-Zr4-Cr-PVD

- The XRD patterns of the substrate Zr-4 and Cr-coated Zr-4 (PVD and Cold spray) confirm the hexagonal closed packed crystal structure of substrate and the presence of Cr layer with cubic crystal structure.
- SEM micrographs evidence a Cr coating thickness of AR-Zr4-Cr-CS and AR-Zr4-Cr-PVD of 29.0±2.0 µm and 6.48±1.41 µm, respectively.

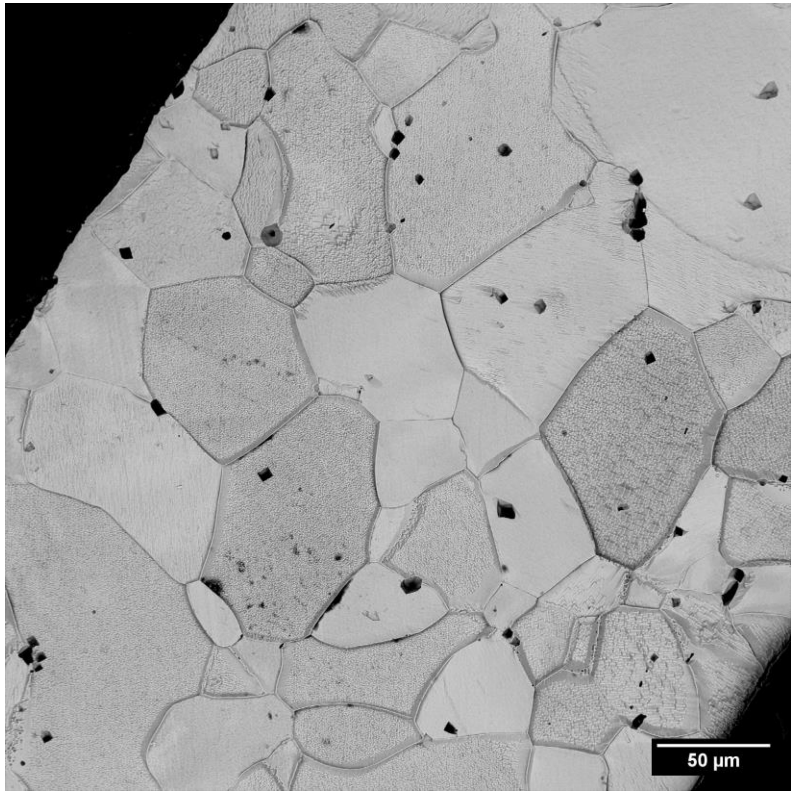


X-ray diffraction patterns of Zircaloy-4 (AR-Zr4), AR-Zr4-Cr-PVD and AR-Zr4-Cr-CS.

FeCrAl Alloys: another alternative



SEM micrograph of etched FeCrAl APMT
Average grain size 8.35 ± 2.03 µm



SEM micrograph of etched FeCrAl C26M
Average grain size 46.04 ± 19.50 µm

%wt.	Cr	Al	Mo	Y	Si	Fe
C26M	11.894	5.263	1.978	0.027	0.279	Balance
APMT	21.146	2.701	3.059	0.127	0.886	Balance

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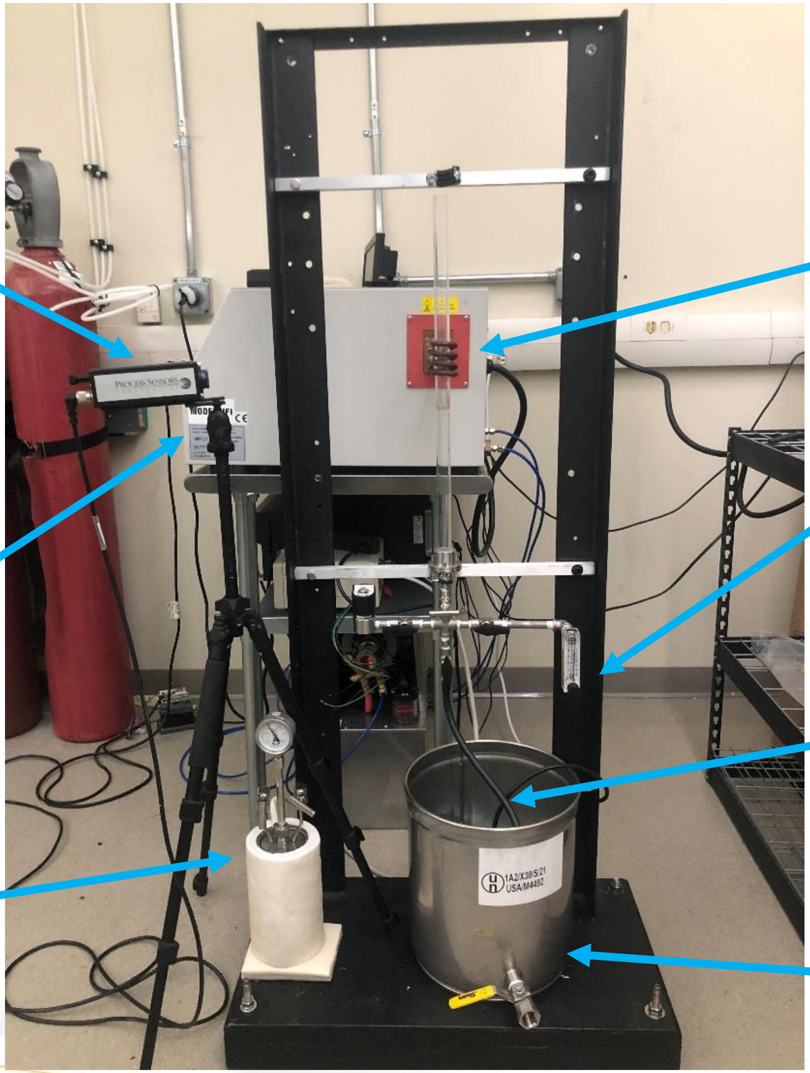
Design of Temperature Excursions and Quenching device

Pyrometer with data acquisition software for measuring temperature during testing

a 7.5 kW high-frequency (100-400 kHz) induction furnace

Stainless steel Bubbler system as steam generator



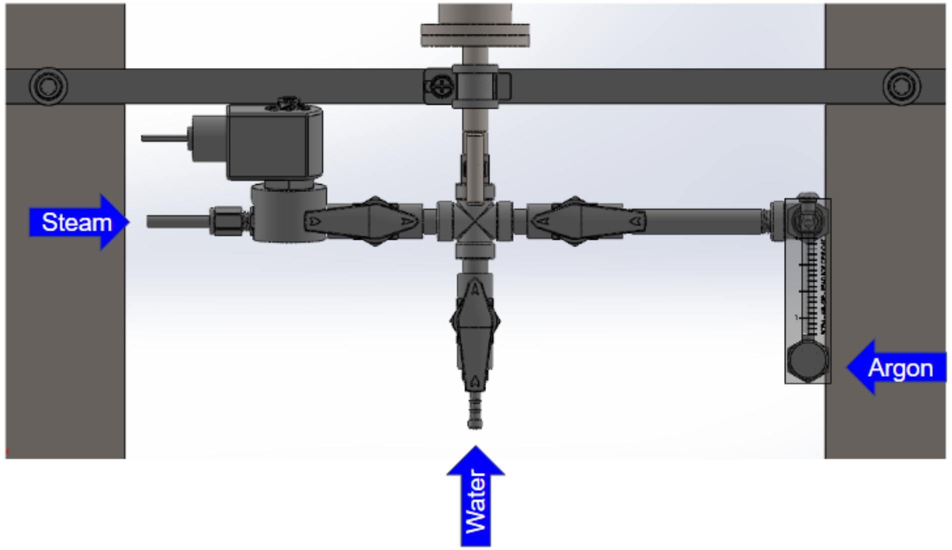
Temperature excursion and quenching setup.

Induction coil where samples are centered for testing

Flowmeter for measuring argon input flow

Submersible Pump for Water, Impact-Resistant/Open-Air, 120V AC

Water container for quenching process



SolidWorks model showing the inputs for steam, water, and argon into the system.

Stainless steel rod

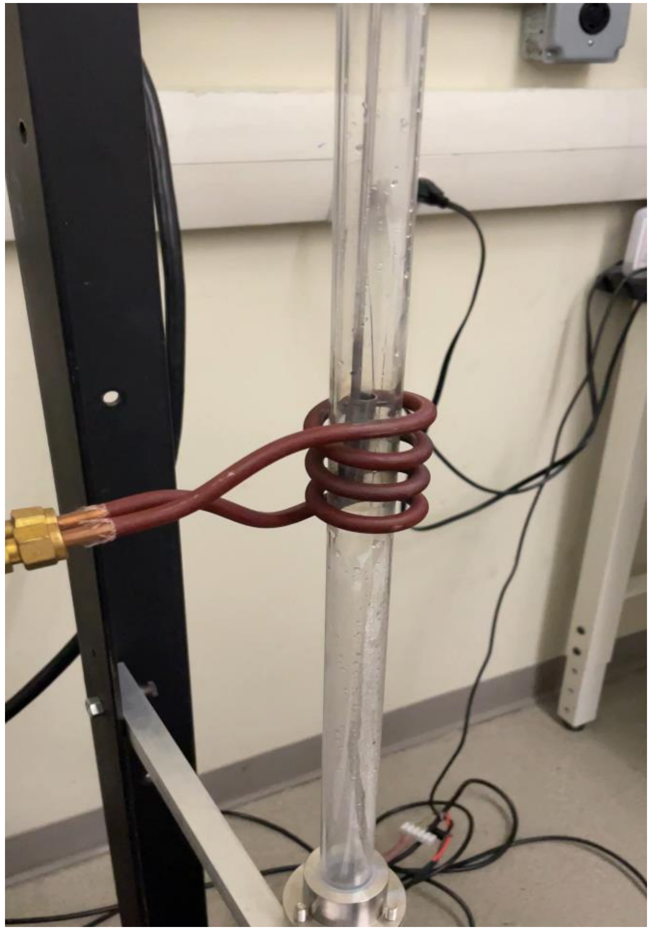
Set screws

Drill bushing

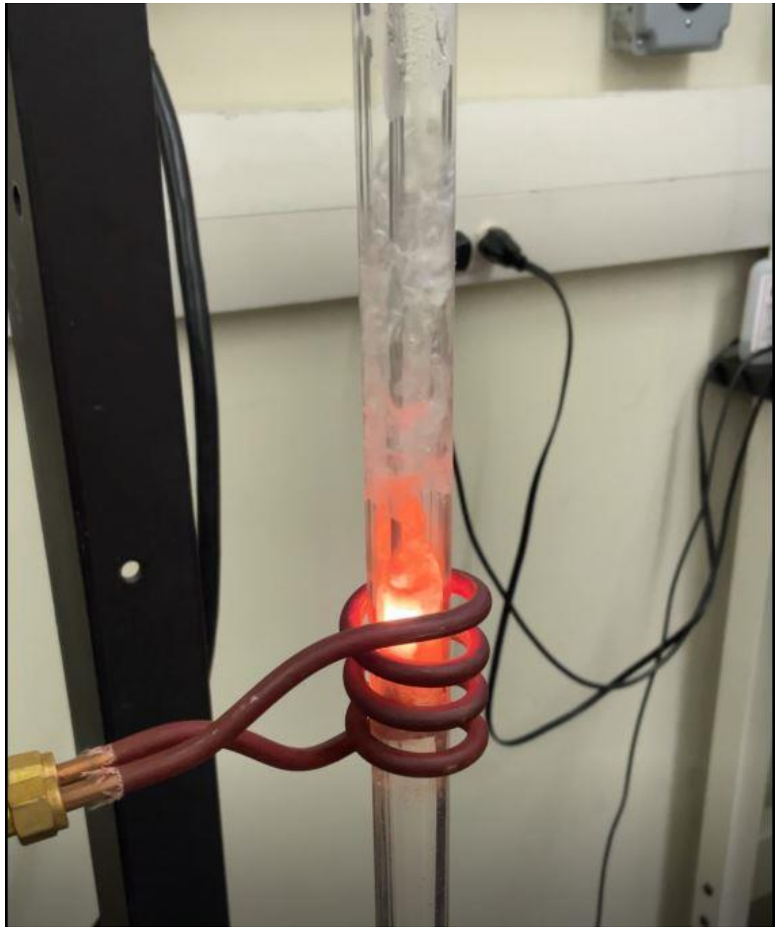
Sample rod

Example of heat excursion and water quenching

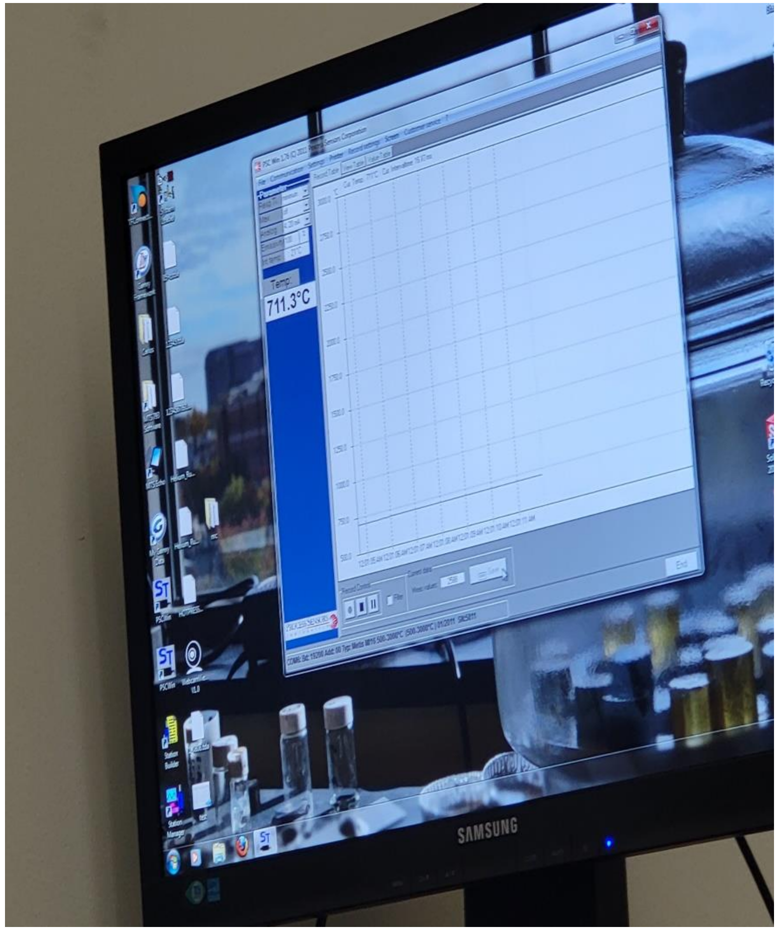
Video of thermal shock event at 30% power of the induction furnace



Photograph of thermal Shock at 30% power of the induction furnace



Date acquisition software recording temperature of the sample from the pyrometer



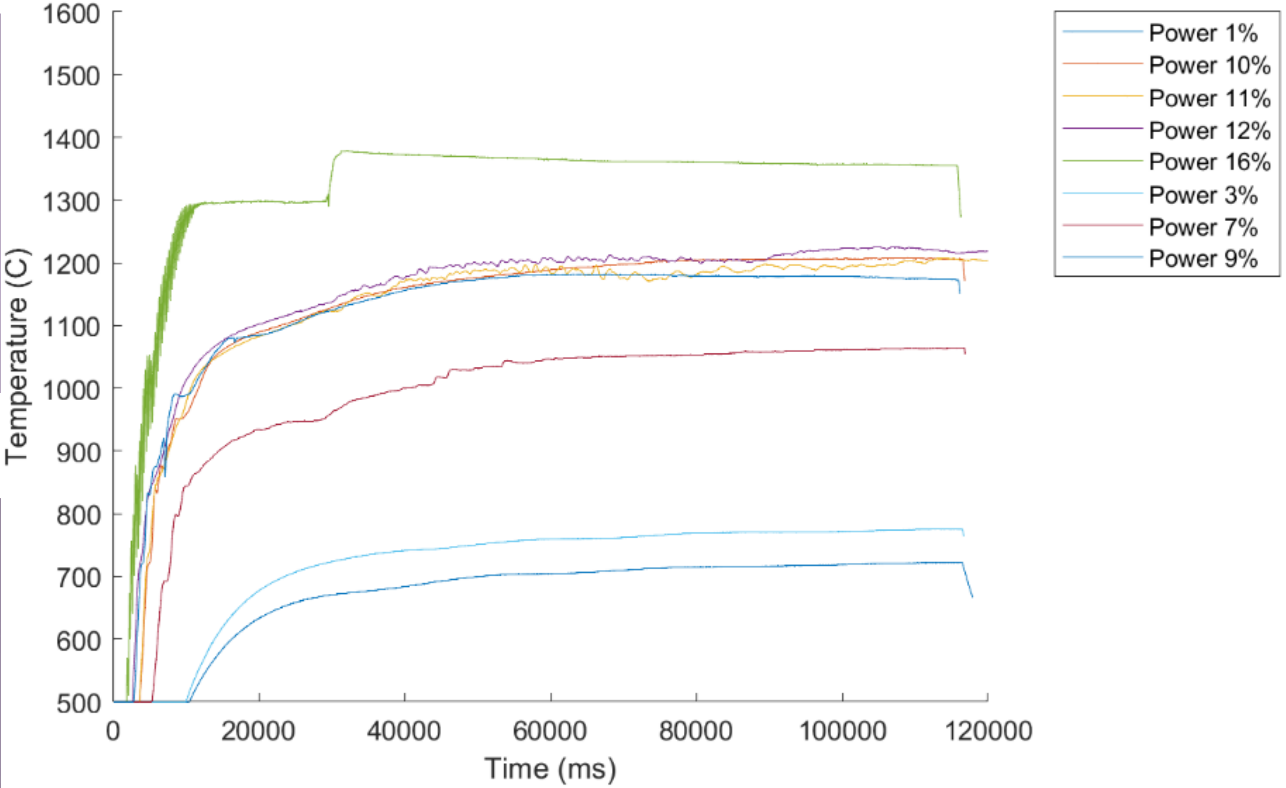
Preliminary results with Zr-4 and Zr-702

1% 5% 10% 16%

5% 10% 15% 20%

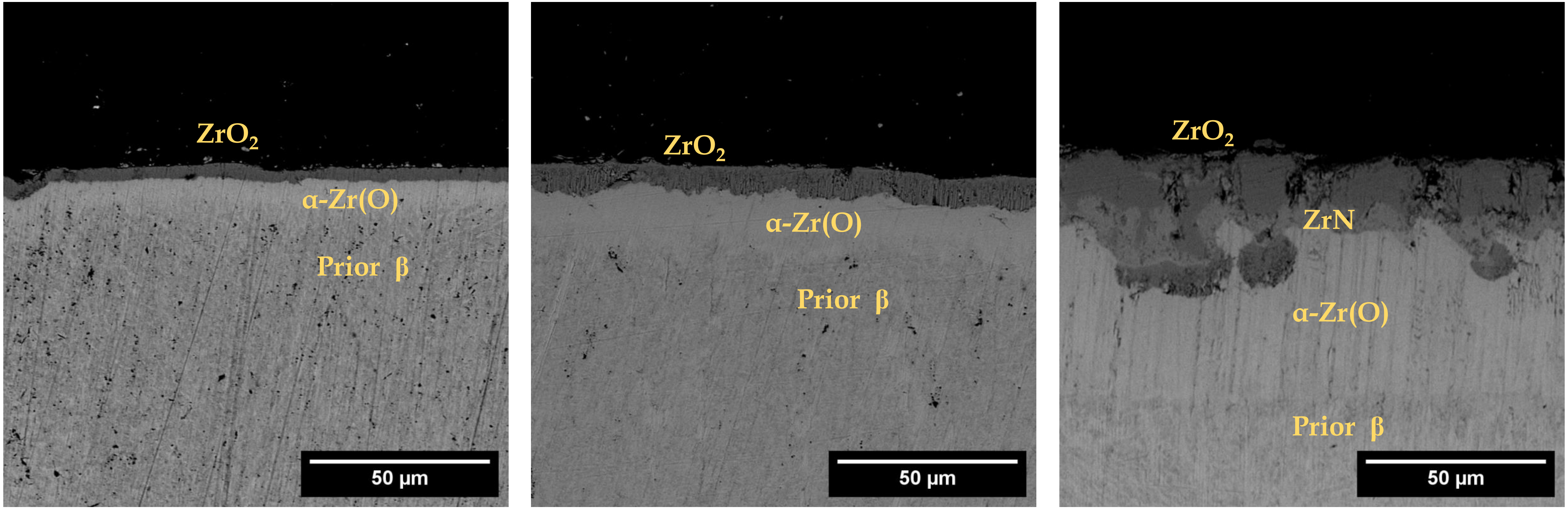
The top images show sample rods heated and cooled in air. The bottom images show sample rods heated with quenching.

Temperature profiles of Zr-4 in dry air



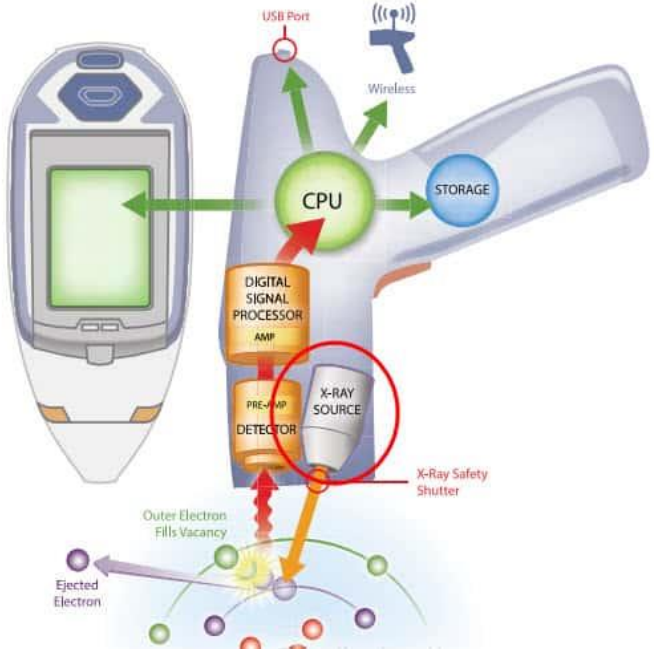
- The system can achieve a maximum heating rate of 100 ºC/s

Preliminary results with Zr-4 and Zr-702

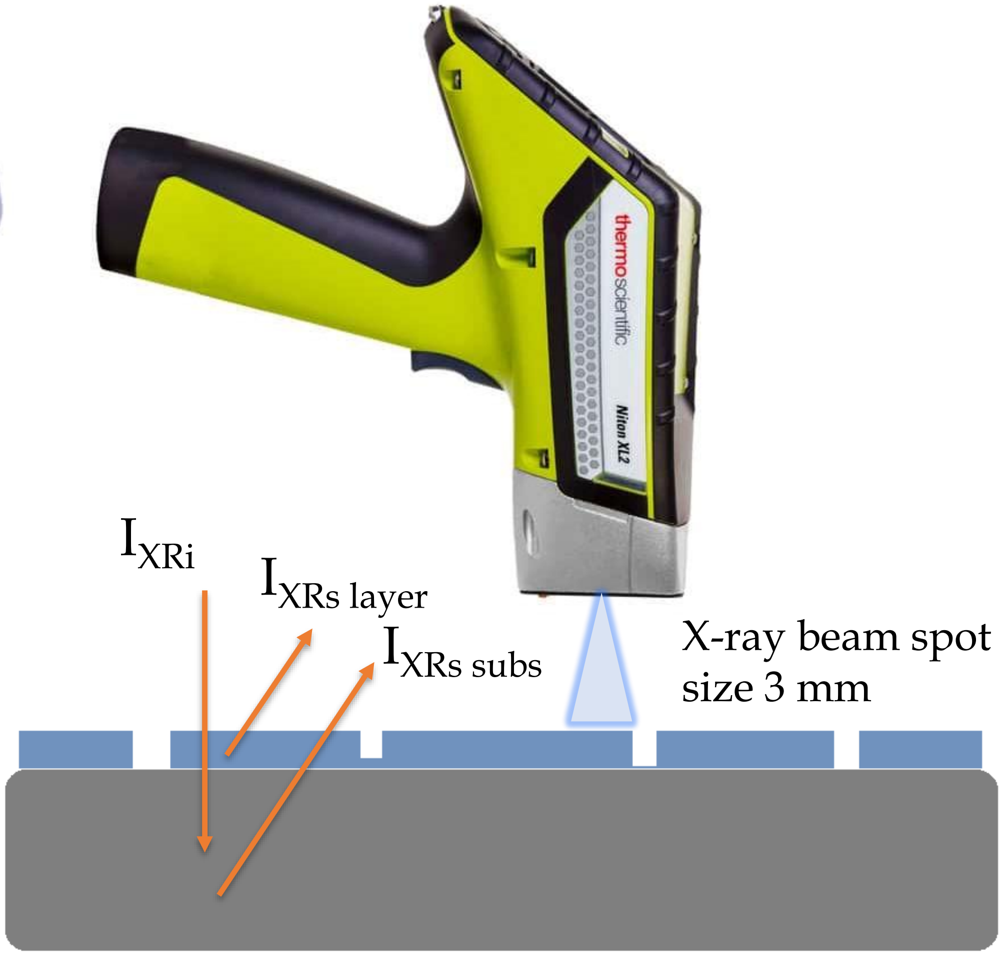


SEM micrographs of Zr-702 after rapid to various temperatures and water quenching a) 800 ºC b) 865 ºC c) 1200 ºC. Average thickness of the oxide layer is 3.4 ± 0.4 µm, 8.8 ± 1.1 µm, 21.9 ± 8.3 µm, respectively

Progress on X-ray fluorescence spectroscopy for ATF Quality control



X-ray fluorescence working principle for elemental analysis



Thickness measurements down to nanometer scale; ability to evidence thickness variations and lack of coating.

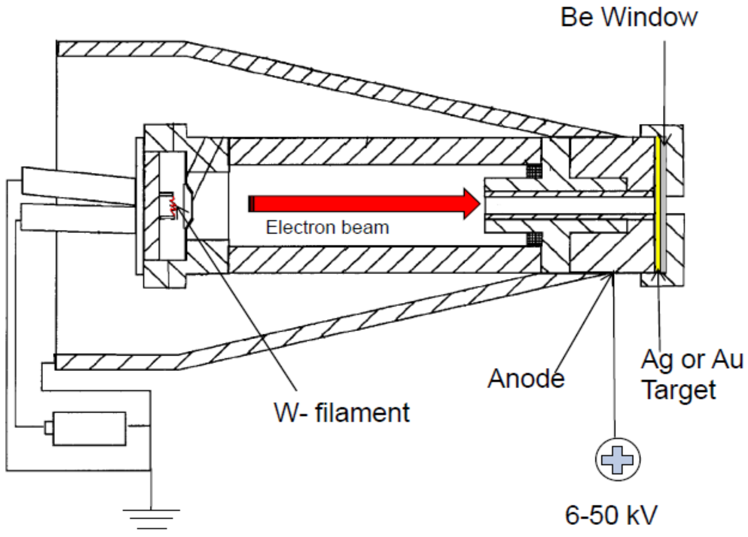
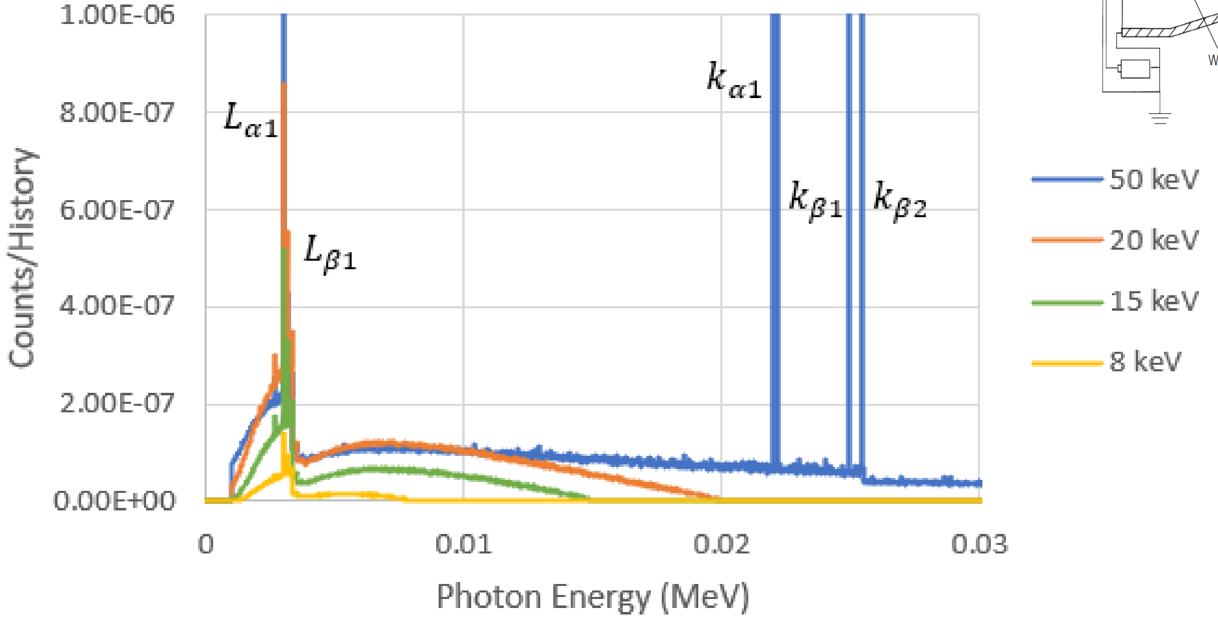
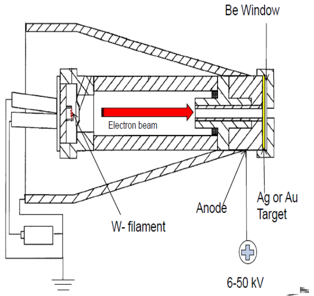
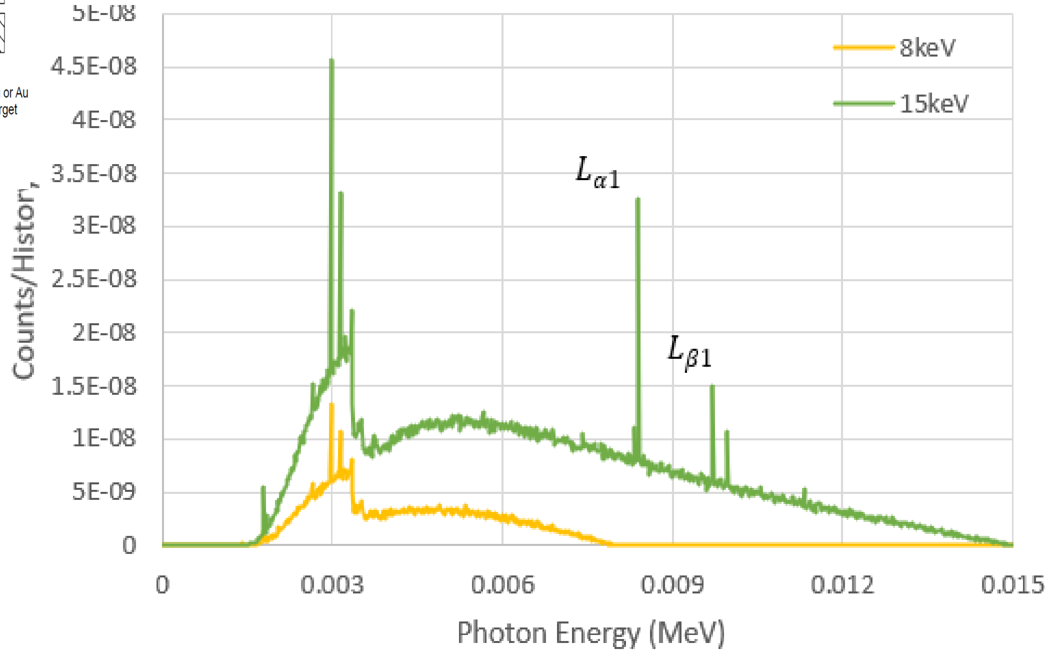


Fig. 1. Diagram of X-ray tube from C. Turner, et al., “Mobile Miniature X-ray Source”

MCNP simulation of the X-ray source for benchmarking with the experimental results







Transmission spectra produced by the Ag target anode with an incident electron beam energy of 8, 15, 20, and 50 keV

Effect of W collimator and 1 µm graphene window on the spectrum generated by an 8 and 15 keV electron beam

- The future work include validating the X-ray simulated beam with the emission spectra from several substrates.
- Defects will be manufactured on the surface of the Cr-coated Zr-4 specimens and XRF spectra at various length steps will be collected to determine the resolution of the device for quality control

Concluding remarks:

- Accident tolerant fuel cladding materials have been acquired and characterized. FeCrAl alloys and Cr-coated Zircaloy-4, produced by PVD and cold spray, are the focus of our work.
- A system for heat excursion and quenching procedures has been designed and built. The system relies the induction mechanism to heat the metallic samples at a high rate. The device includes several components that includes gas, steam, and water inlets.
- Preliminary tests have been conducted with Zr-4 and Zr-702 to optimize the device. The results obtained for these specimens aligned with those reported in the literature on high temperature oxidation of Zircaloys in various environments.
- Monte Carlo N-Particle simulations have been completed to generate an X-ray spectrum of our handheld device. The results of the modeling implemented to achieve the output spectrum of the XRF device are supported by the presence of predicted characteristic X-rays and X-ray attenuation.

Future work:

- Design of experiments for FeCrAl and Cr-coated Zr cladding for high temperature excursions and quenching.
- The output spectra obtained for the XRF device will be used as the source definition for future models. The spectra will be used to produce fluorescence in various material specimens.
- Dry storage simulated environments, temperature profiles and hoop stresses, are being determined. A loading mechanism has been designed and is being manufactured.

Thank You!!

Questions??

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