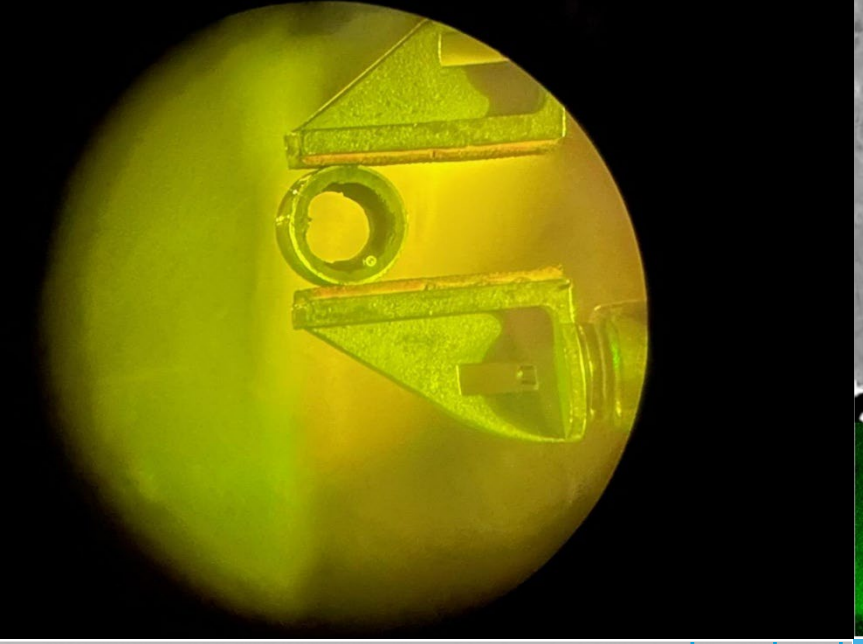
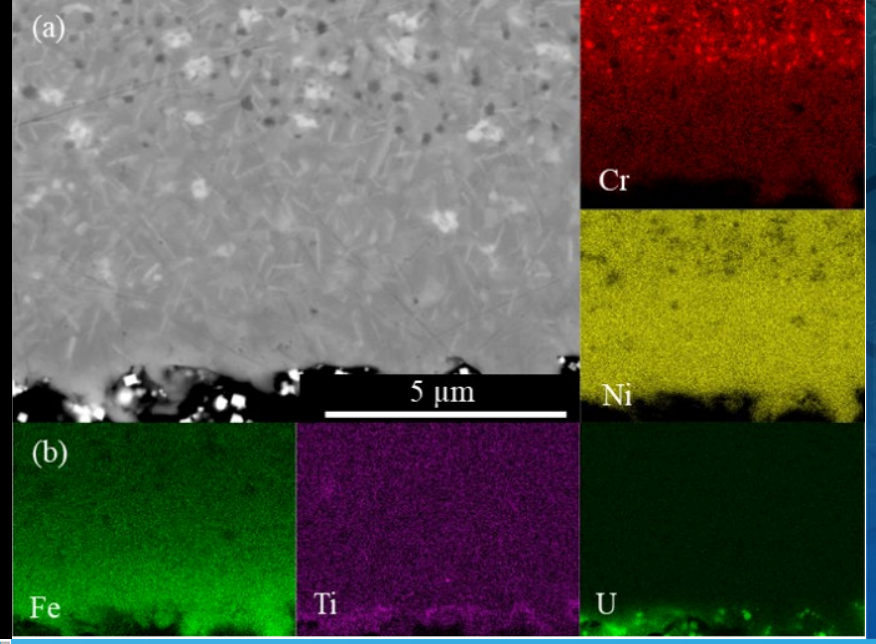
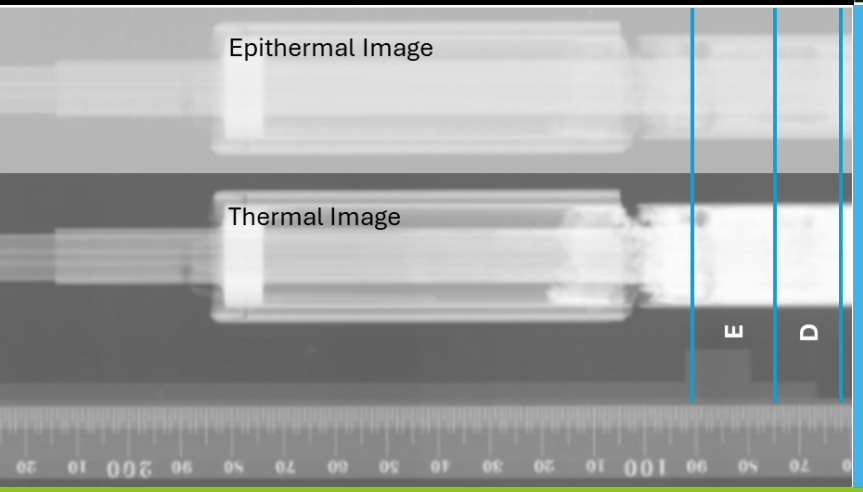





Jaewoo Park, Tammy Trowbridge, Austin Poole, Cassie Anderson-Thueson, Steven Pappas, David Zirker, Jakeob Maupin, Brian Kajganich, Marc Babcock, Katie Hawkins, Glen Papaioannou, Nick Erfurth, Tony Jones, Matthew Arrowood, William Phillips, Chuting Tsai, Calvin Downey, Morgan Kropp, Stephen Warmann, Abdalla Abou-Jaoude, and Toni Karlsson




# Molten Salt Temperature – Controlled Irradiation (MRTI)

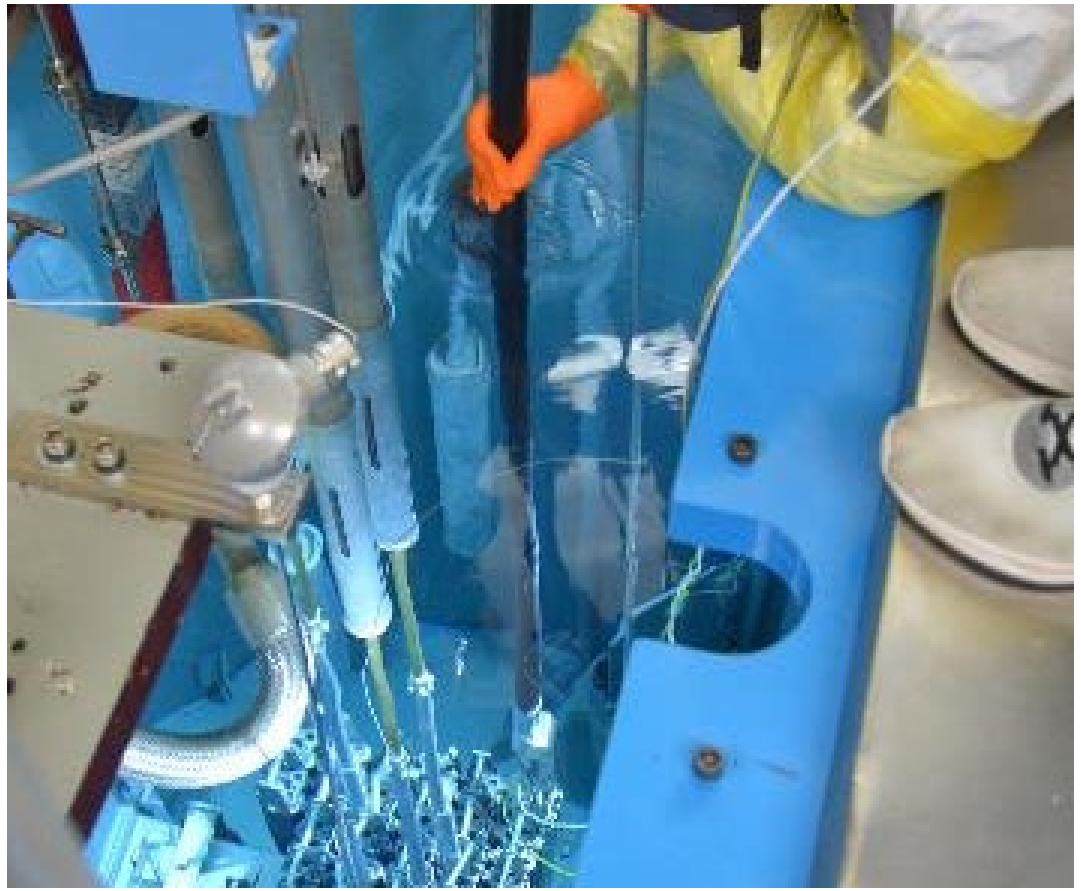
## Project Summary

Battelle Energy Alliance manages INL for the
U.S. Department of Energy's Office of Nuclear Energy

Idaho National Laboratory

# Introduction

- Purpose - Establish of a domestic neutron irradiation and characterization capabilities for fissile material-bearing salts for Molten Salt Reactor (MSR) RD&D and assess fuel and structural material performance
- Focus
  - Radioactive Source Term Quantification
  - Thermophysical Property Evolution
  - Salt-facing Materials Corrosion



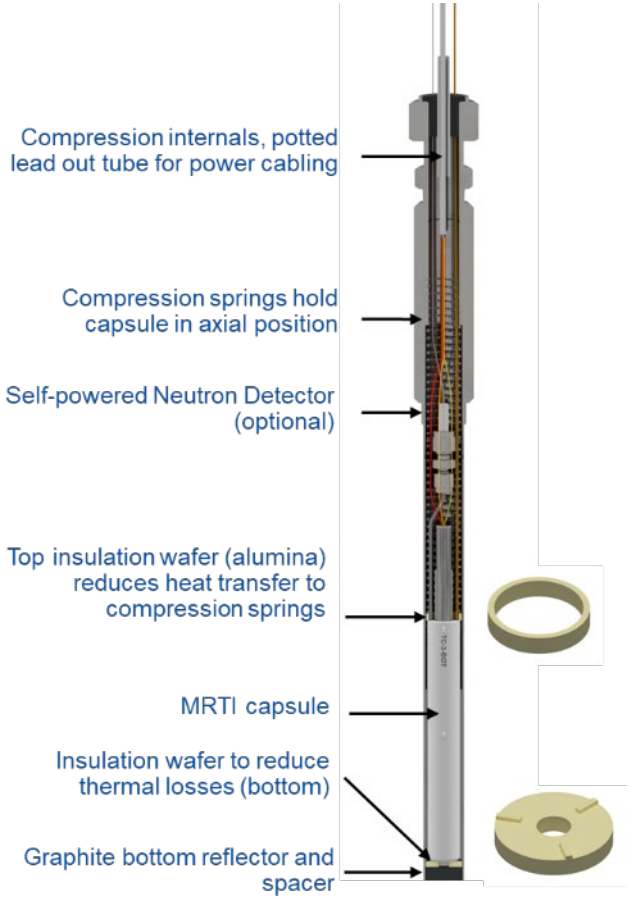
# MRTI - Overview

- Salt: ~41g of $33UCl_3$-67NaCl (93% $^{235}U$)
- Capsule material: Inconel-625
- Other structural material: SS-316
- Plenum gas: Ar
- Outer can gas: 15He/85Ar
- Predicted fuel performance under irradiation
  - Fission Heat = 20 $W/cm^3$
  - Neutron Flux = $3.5x10^{12}$ $n/cm^2$-s
  - Gamma Flux = $1.4x10^{13}$ $\gamma/cm^2$-s
  - Salt Temperature = 525-900°C

SS = Stainless Steel

# MRTI – Overview

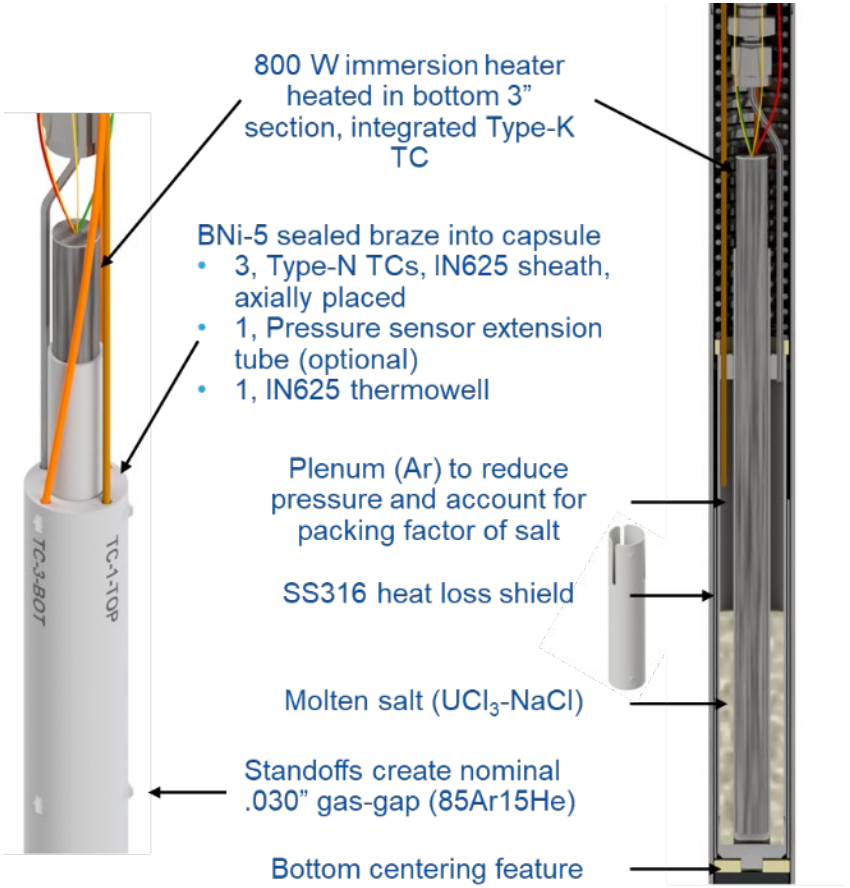
## Outer Can/Cluster




TC = Thermocouple

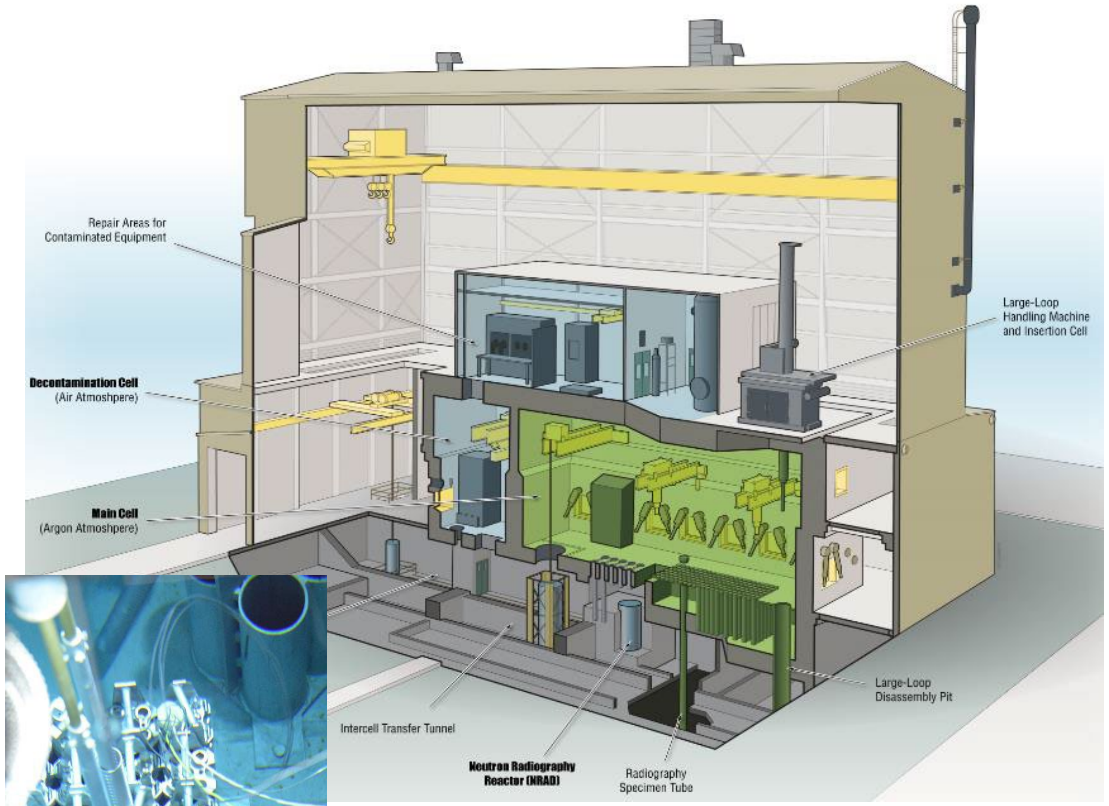
IN625 = Inconel 625

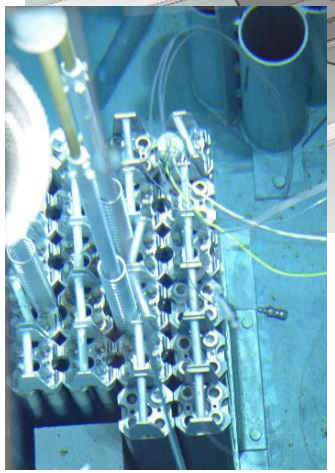
## Inner Capsule



# MRTI - Overview

- Neutron RADiography (NRAD) Reactor
- 250 kW TRIGA-fuel MTR-grid pool reactor
  - Typically used for neutron radiography PIE
  - Pool reactor (no pressure)
  - Only TRIGA that handles HEU
  - No experiment traffic/schedule, irradiate by request
- Immediate access to HFEF for quicker turnaround PIE

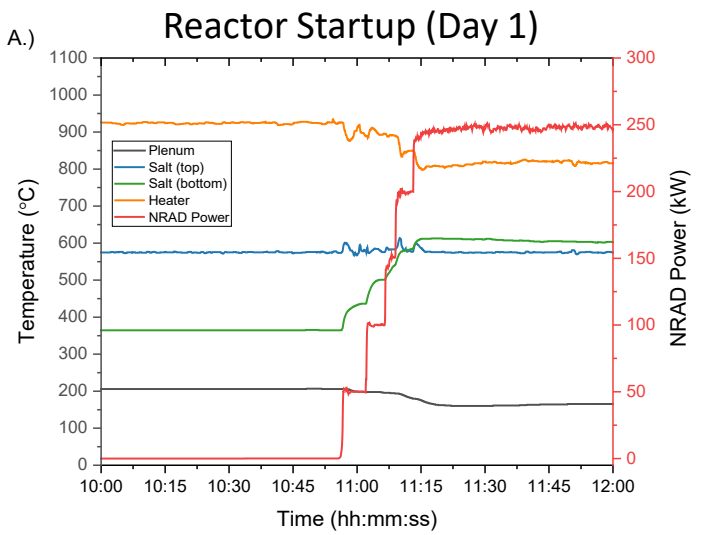


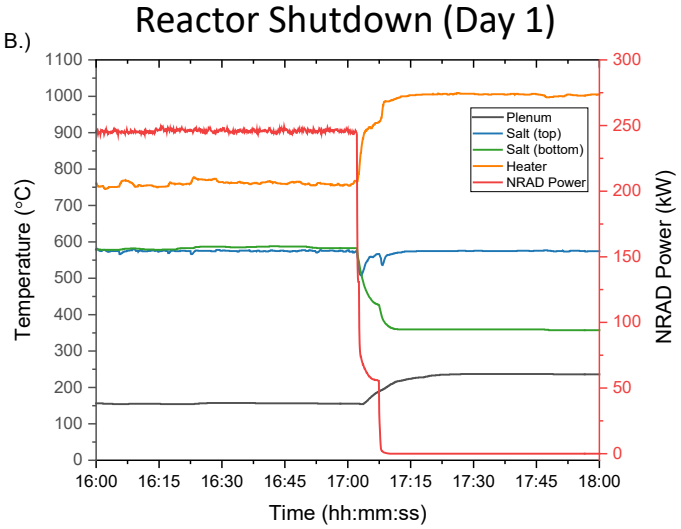



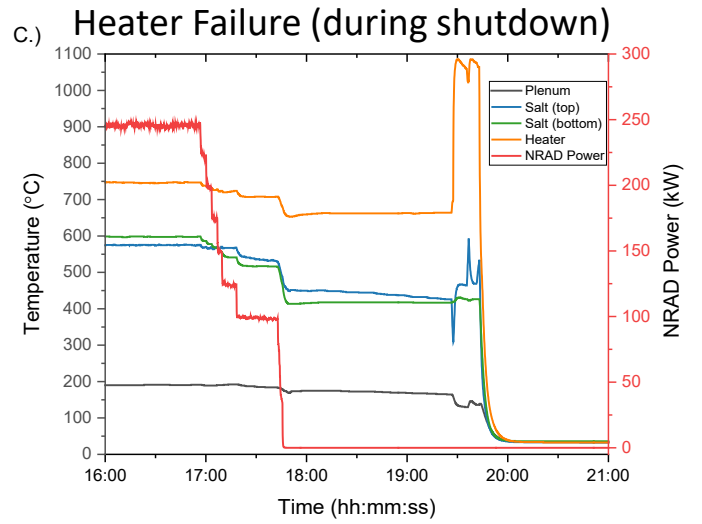
NRAD = Neutron RADiography
TRIGA = Training, Research, Isotope, General Atomics
MTR = Materials Test Reactor
HEU = Highly Enriched Uranium
HFEF = Hot Fuel Examination Facility

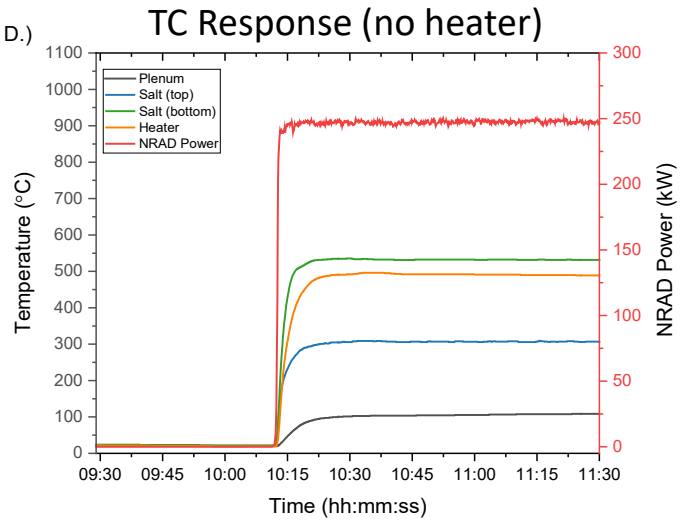
# MRTI - Irradiation

- Irradiation started on August 21st 2023
- Final irradiation on June 3rd 2024
  - 390 hr of irradiation
  - 0.20 GWd/MTU burnup
- Transferred into HFEF main cell on June 25th 2024
- Disassembled in July 2024



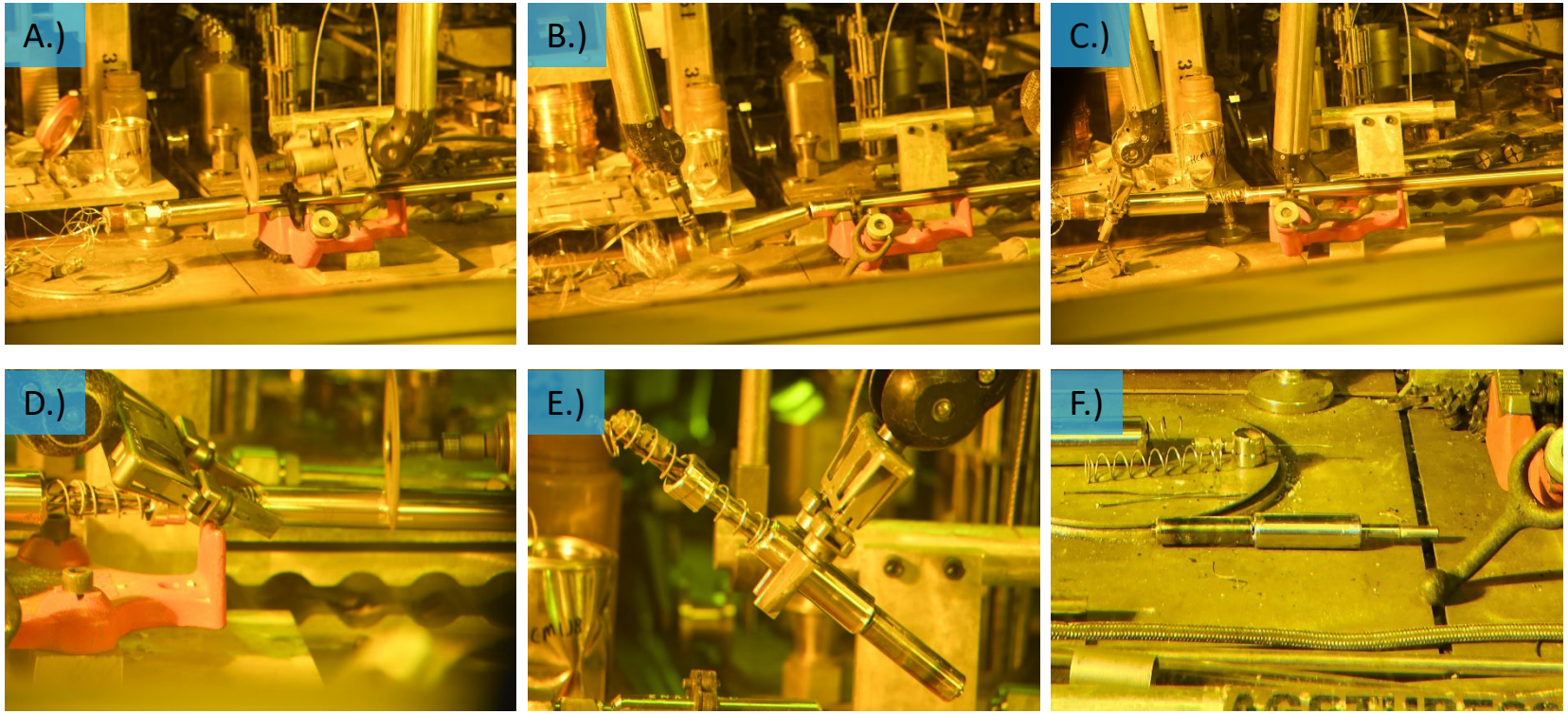




GWd = Gigawatt days

MTU = Metric Ton of Uranium

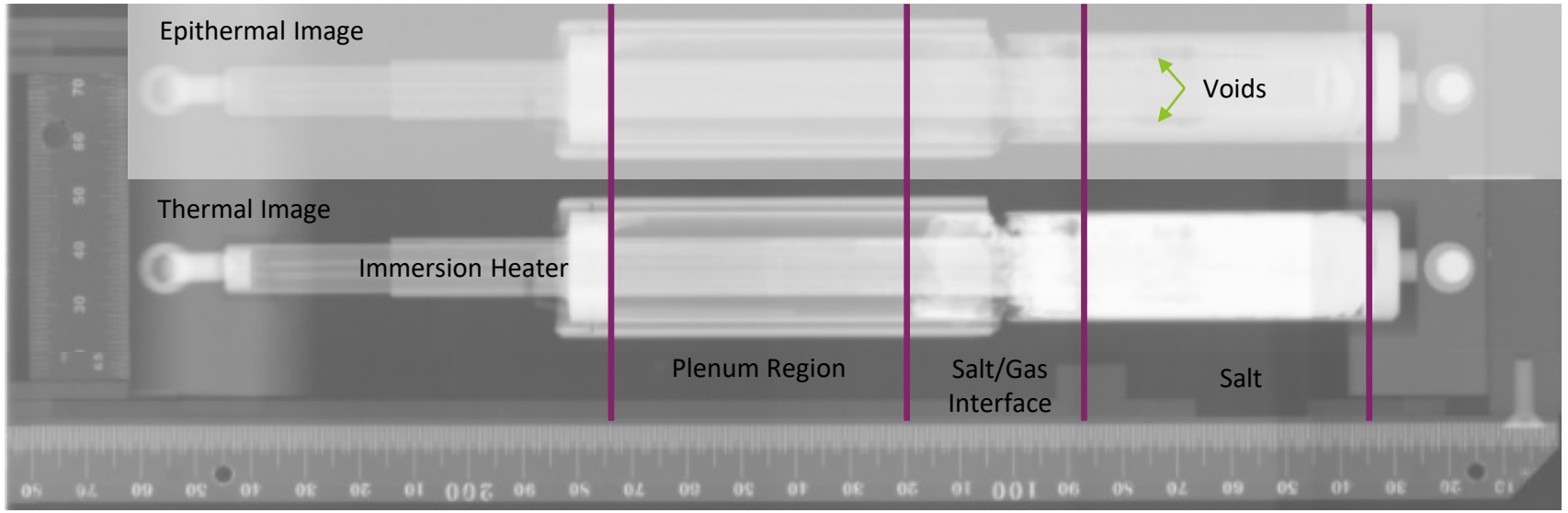
# MRTI – PIE

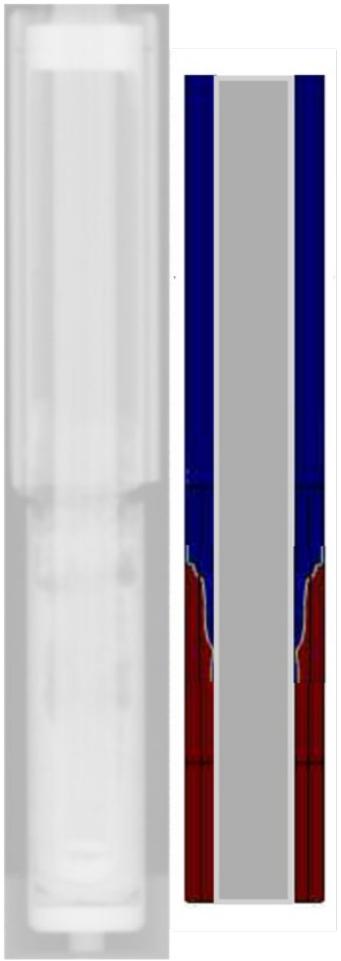
- Remotely disassembled in Ar hot cell
- Die grinder used to remove capsule from outer can
- Capsule was breached during the process, unable to collect off-gas sample
- Immersion heater “stuck” in place



# MRTI - PIE

- Neutron radiograph imaging
  - 5 angles: 0, 45, 90, 120, 240
  - Thermal and epithermal imaging
    - Epithermal neutrons have higher energy, can penetrate further into the sample than thermal neutrons
    - Solidification modeling using CFD

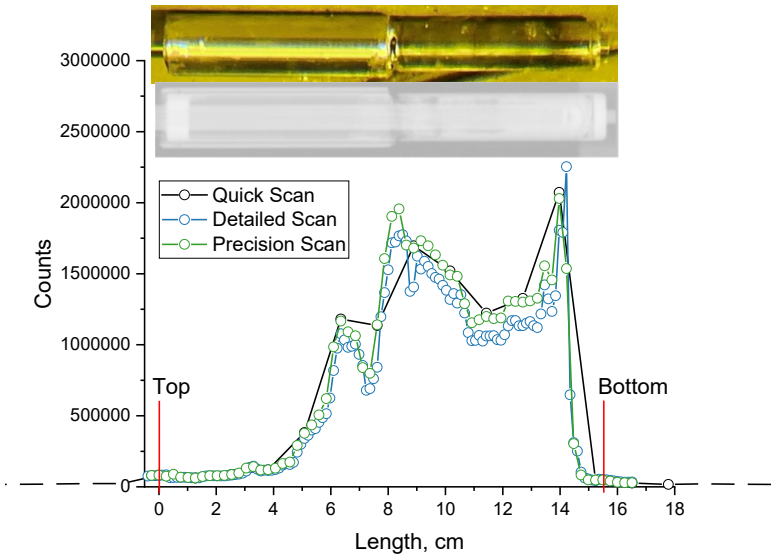


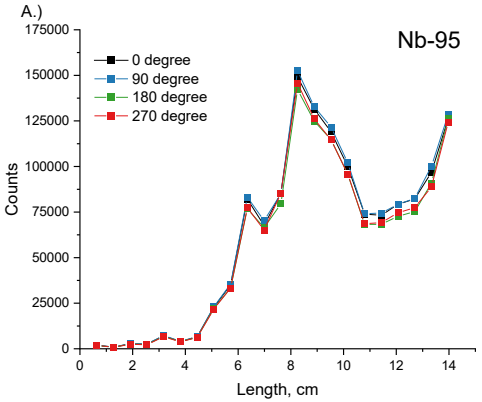



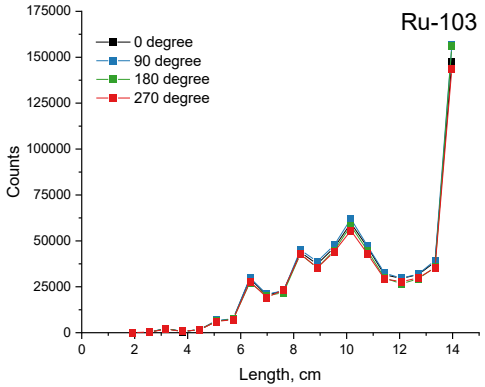
CFD = Computational Fluid Dynamics

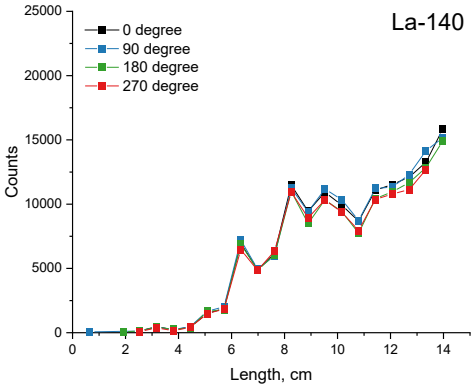
# MRTI - PIE

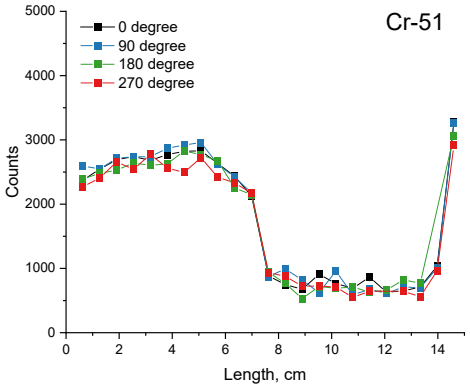
- PGS the length of the capsule
  - Useful energy range ~150eV – 1600keV
  - Background, quick, detailed scans (4 angles)
- Expected FP (ORIGEN 2 software):
  - $Nb^{95}$, $Zr^{95}$, $Y^{91}$, $Ce^{141}$, $Sr^{89}$, $Ru^{103}$, $Pr^{144}$, $Ce^{144}$, $Pr^{143}$, $La^{140}$, $Ba^{140}$, $S^{35}$, $Nd^{147}$, $Ru^{106}$, and $Rh^{106}$
- Activated material of construction
  - $Cr^{51}$, $Co^{60}$, etc. (low counts)



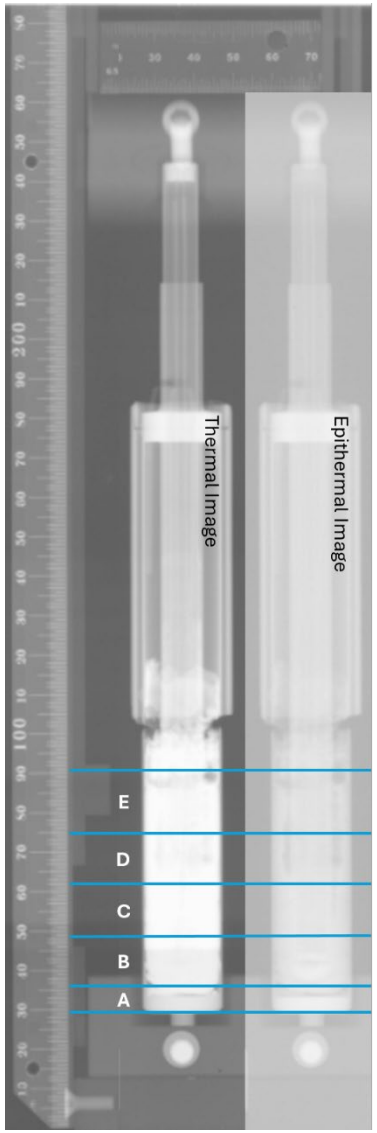

PGS = Precision Gamma Scan

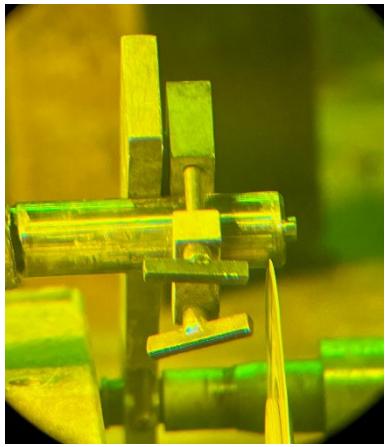
keV = kilo- electron volts

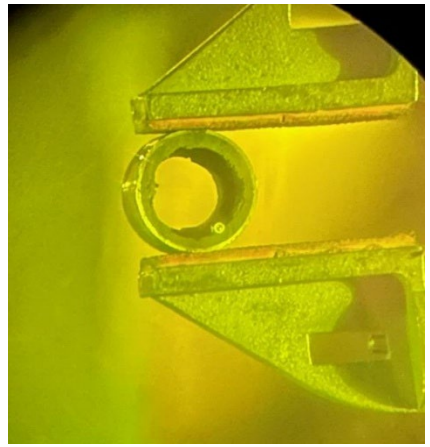
FP = Fission Product

# MRTI - PIE

- MRTI sectioning
  - Low speed saw
  - Analysis of Segment E
    - SEM (material of construction)
    - Elemental/isotopic (salt)
  - Analysis of Segment B
    - Elemental/isotopic (salt)
  - Salt melted out of segment
  - All other segments/salt are stored in individual capsule in HFEF
  - Two slices extracted from Segment E
    - Radial - mounted in epoxy
    - Planar - mounted on SEM ‘sticky dot’



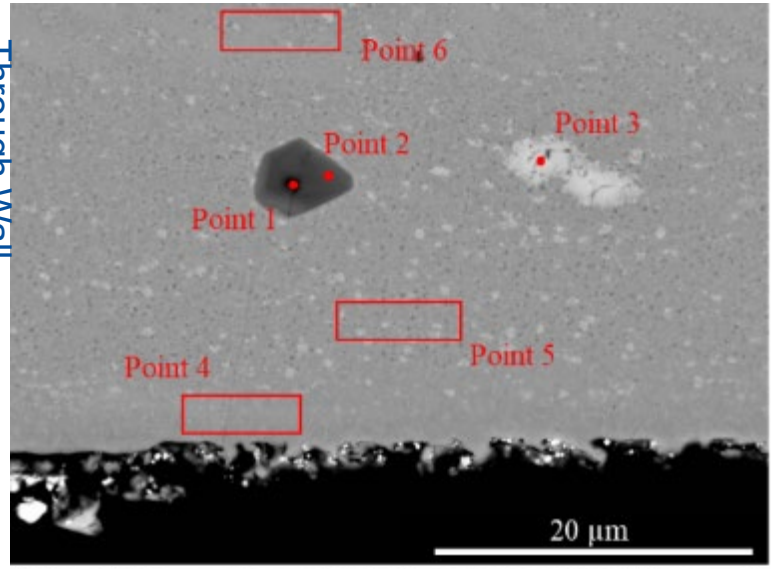





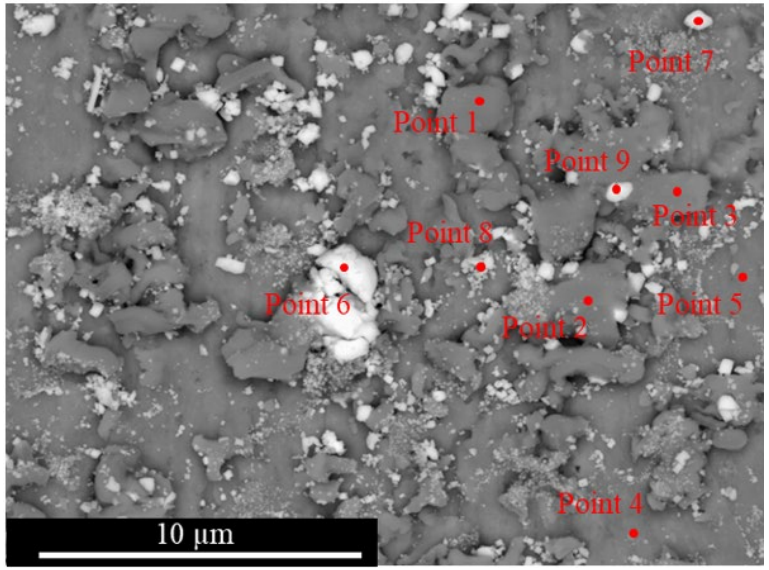
SEM = Scanning Electron Microscope

# Microscopy

Through Wall




Facing Wall




- BSE images show clear corrosion of the IN625 capsule inner surface with initial pitting, and composition changes from redox reactions during high temperature irradiation.
- Segregation of Ti and Nb into precipitates within IN625 due to raw material processing or time-at-temperature during irradiation.

| Point | Concentration, wt.% | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | Cr | Fe | Ni | Mo | Nb | Ti | N | Mg | Al |
| 1 | 2.14 | 0.00 | 1.31 | 0.00 | 14.92 | 56.23 | 12.52 | 10.08 | 2.80 |
| 2 | 2.67 | 0.00 | 1.82 | 0.00 | 23.69 | 60.09 | 11.74 | 0.00 | 0.00 |
| 3 | 1.28 | 0.00 | 3.33 | 0.00 | 92.27 | 3.12 | 0.00 | 0.00 | 0.00 |
| 4 | 18.25 | 7.34 | 63.24 | 6.79 | 4.39 | 0.00 | 0.00 | 0.00 | 0.00 |
| 5 | 22.65 | 3.86 | 59.60 | 9.05 | 4.84 | 0.00 | 0.00 | 0.00 | 0.00 |
| 6 | 22.82 | 3.83 | 59.88 | 8.71 | 4.76 | 0.00 | 0.00 | 0.00 | 0.00 |

- Precipitates with elevated Ti, Nb, N suggest (Nb,Ti)C and TiN, δ-phase $Ni_3Nb$.
  - Formation from high temperature during ~400hour operation
- Cr depletion at point 4 versus point 5

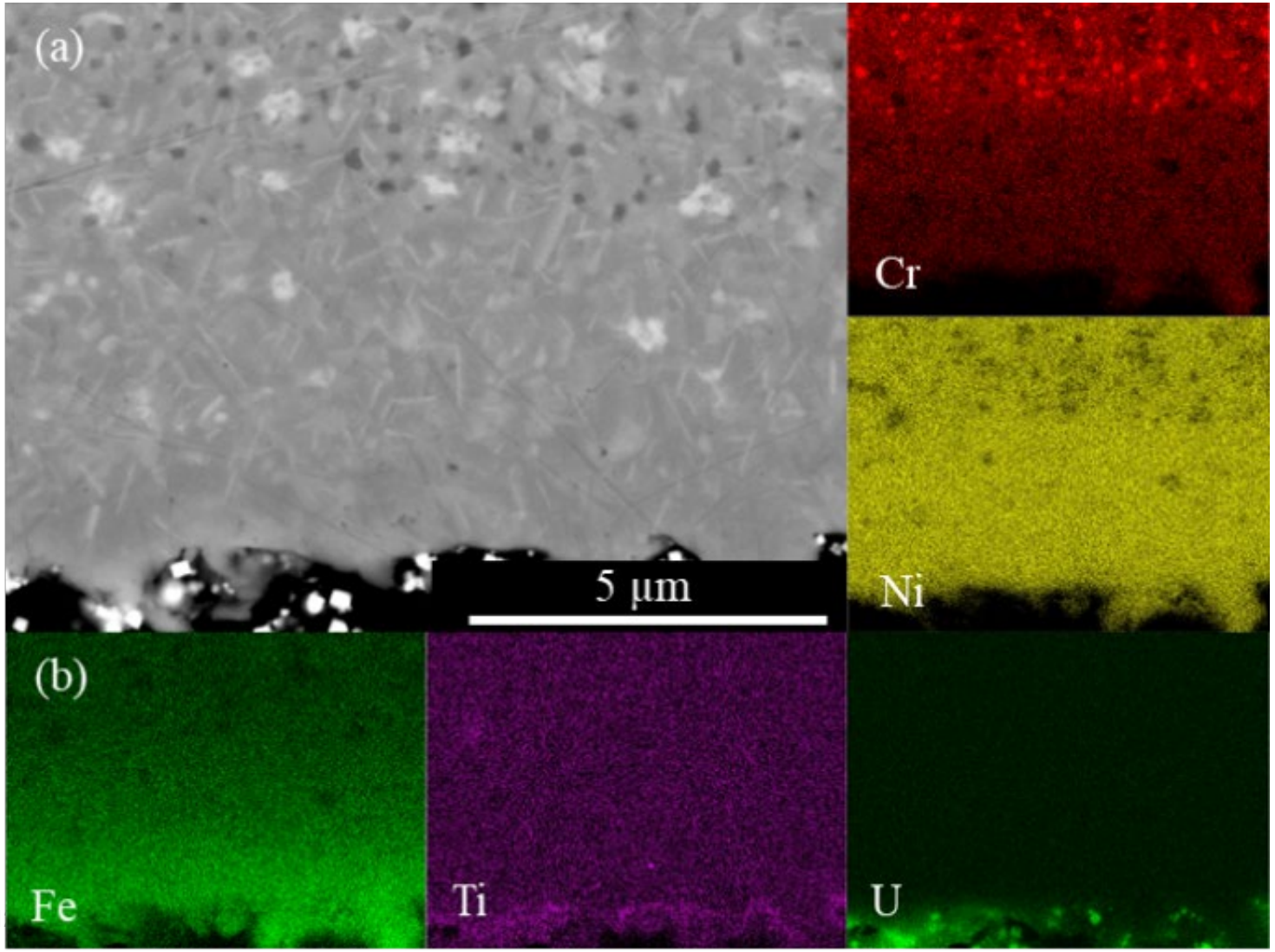
| Point | Concentration, wt.% | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | Cr | Fe | Ni | Mo | Nb | U | O | Al | Si | Ti |
| 1 | 20.01 | 13.20 | 61.92 | 2.93 | 1.65 | 0.00 | 0.00 | 0.00 | 0.28 | 0.00 |
| 2 | 19.80 | 12.39 | 60.42 | 2.15 | 1.30 | 3.42 | 0.00 | 0.00 | 0.22 | 0.31 |
| 3 | 18.89 | 11.88 | 58.36 | 3.57 | 2.00 | 4.14 | 0.00 | 0.18 | 0.35 | 0.63 |
| 4 | 18.67 | 9.59 | 58.42 | 7.37 | 3.59 | 0.79 | 0.00 | 0.00 | 0.77 | 0.80 |
| 5 | 18.18 | 10.53 | 59.36 | 6.46 | 3.25 | 0.93 | 0.00 | 0.00 | 0.78 | 0.51 |
| 6 | 2.09 | 1.00 | 5.39 | 0.08 | 0.49 | 77.96 | 12.87 | 0.00 | 0.12 | 0.00 |
| 7 | 5.90 | 2.70 | 13.47 | 1.46 | 2.16 | 57.28 | 16.27 | 0.00 | 0.39 | 0.37 |
| 8 | 16.12 | 8.68 | 45.03 | 3.88 | 2.25 | 22.97 | 0.00 | 0.00 | 0.46 | 0.61 |
| 9 | 11.75 | 8.57 | 27.72 | 0.68 | 1.04 | 49.94 | 0.00 | 0.00 | 0.30 | 0.00 |

- Uranium (U) and UO2 observed on surface of IN625 capsule wall

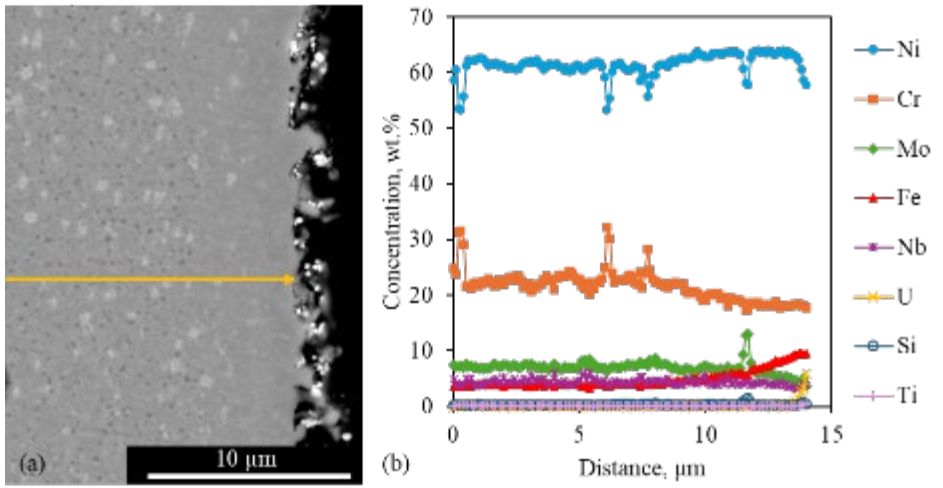
EDS = Energy Dispersive X-Ray Spectroscopy

# Microscopy

**SEM-EDS** A.) SEM image of IN625 capsule surface and B.) SEM-EDS mapping/line scan for Cr, Ni, Fe, Ti, and U.




- Cr and Mo were depleted at the corrosion surface to a depth of approximately 6 µm. The concentration of Fe and Ni was increased.
  - $MCl_2 + Cr \rightleftarrows 3CrCl_2 + M$; M= Ni, Fe
- Uranium metal deposits detected on the surface of the IN625.
  - $4U^{3+} \rightleftarrows 3U^{4+} + U$
  - Can occur with disproportionation of $U^{3+}$ and the fission of U235 during irradiation. This may also result in decreased concentration of $UCl_3$.
- Assuming a 5 µm corrosion layer and 390 hours of irradiation time, the corrosion rate is calculated to be 0.11mm/year, a relatively low corrosion rate



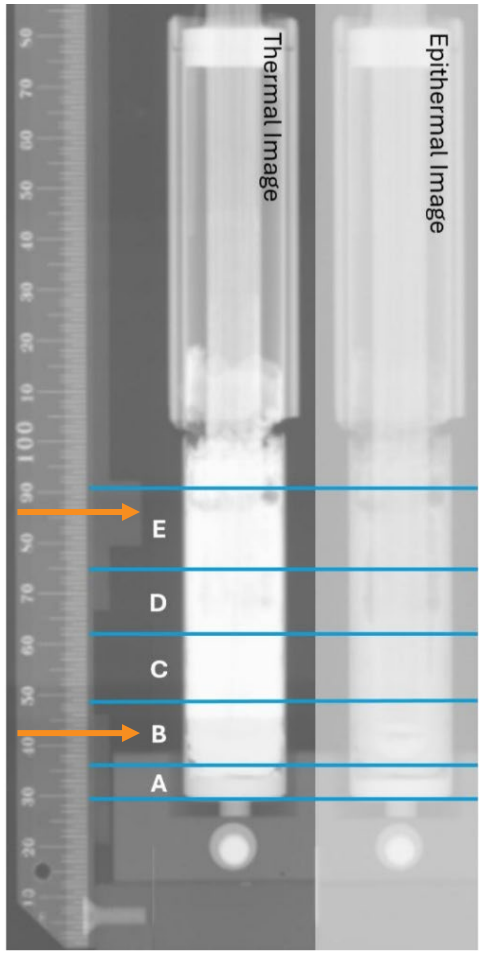
# Chemical Analysis

- ICP-MS and ICP-OES revealed differences in molten salt chemistry and FP inventory after irradiation.
  - No clear conclusion on FP stratification due to cycling (but it is happening!).

| Element | Concentration, µg/g (Error, %) | |
|---|---|---|
| | Segment B | Segment E |
| U-234 | 4940 (5) | 4720 (5) |
| U-235 | 465000 (5) | 449000 (5) |
| U-236 | 1240 (5) | 1190 (5) |
| Np-237 | 0.0616 (25) | <0.03 (N/A) |
| U-238 | 29100 (5) | 27900 (5) |
| Pu-239 | 4.52 (15) | 2.26 (10) |
| Pu-240 | 1.17 (20) | 0.454 (10) |
| Zr-91 | 2.5 (10) | 2.47 (10) |
| Nb-93 | 5.82 (15) | 1.45 (15) |
| Ru-100 | 7.61 (10) | <2 (N/A) |
| Ru-101 | 3.53 (10) | <4 (N/A) |
| Ru-102 | 3.14 (30) | <0.6 (N/A) |
| Ru-104 | <3 (N/A) | <3 (N/A) |
| Cs-133 | 13.6 (5) | 5.37 (10) |
| Cs-135 | 2.79 (10) | 2.06 (5) |
| Ce-140 | 4.77 (10) | 5.06 (10) |
| Ce-142 | 4.26 (10) | 4.42 (10) |
| La-139 | 4.86 (10) | 4.61 (15) |

| Element | Radioactivity, µCi/grams (Error, %) | |
|---|---|---|
| | Segment B | Segment E |
| Am-241 | <2 (N/A) | <1 (N/A) |
| Ce/Pr-144 | $4.16 \times 10^3$ (3) | $4.69 \times 10^3$ (3) |
| Ce-141 | 22.0 (4) | 27.0 (3) |
| Co-58 | 0.850 (17) | <0.4 (N/A) |
| Co-60 | 0.765 (8) | <0.3 (N/A) |
| Cs-134 | <0.3 (N/A) | <0.5 (N/A) |
| Cs-137 | $3.12 \times 10^2$ (3) | $2.27 \times 10^2$ (3) |
| Eu-154 | <0.7 (N/A) | <1 (N/A) |
| Eu-155 | 9.02 (6) | <2 (N/A) |
| I-131 | <0.5 (N/A) | <0.6 (N/A) |
| Mn-54 | <0.1 (N/A) | <0.4 (N/A) |
| Nb-95 | $7.93 \times 10^2$ (3) | $2.34 \times 10^2$ (3) |
| Ru/Rh-106 | $3.24 \times 10^2$ (3) | 39.5 (10) |
| Ru-103 | 45.3 (3) | 4.75 (4) |
| Sb-125 | 24.9 (3) | <2 (N/A) |
| Y-91 | $4.04 \times 10^2$ (16) | $3.65 \times 10^2$ (10) |
| Zr-95 | $3.84 \times 10^2$ (3) | $1.15 \times 10^2$ (3) |

- Partially solidified after heater failure.
- Molten during entire irradiation.



# Conclusions

- The MRTI experiment irradiated 13 cm³ of $UCl_3$-NaCl fuel salt (93% U-235), with a burnup of 0.196 GWd/MTU over 390 hours
- First irradiation study of enriched uranium chloride fuel salt examining corrosion interactions with structural alloy under neutron irradiation conditions
- PGS and neutron radiography, helped visualize solidification and the distribution of gamma-emitting fission products
- Noble metal fission products (Rh, Ru, Sb, Tc, Te) concentrated at bottom while soluble lanthanides remained uniformly distributed throughout salt
- Corrosion products including Fe, Ni, Cr, and Mn showed higher concentrations in bottom salt due to active dissolution and limited diffusion
- Cr and Mo depletion (~5 μm depth) with Fe and Ni enrichment observed near capsule surface due to reduction-oxidation reactions
- Corrosion rate of 0.11mm/year
- Heater failure created temperature gradient causing salt stratification with higher NaCl concentration at top and increased fission products, especially the noble metals at bottom

Irradiation of an Enriched Uranium (NaCl-$UCl_3$) Fuel Salt Capsule, Summary of Nondestructive Post Irradiation Examinations, and Solidification Modeling

Toni Karlsson[1*], Abdalla Abou-Jaoude[1], Ramiro Oscar Freile[1*], Morgan Kropp[1], Steve Warmann[1], Evan Lovel[1], Brian Kajganich[1], Marc Babcock[1], Katie Hawkins[1], Calvin Downey[1], Mauricio Tano Retamales[1], William Phillips[1], Glen Papaioannou[1], Chuting Tsai[1], Michael Ruddell[1], Reed Eichele[1], Steven Pappas[1], Charles Pierce Jones III[1], Richard M. Cox[2], and Patricia Paviet[2]


*Battelle Energy Alliance manages INL for the U.S. Department of Energy's Office of Nuclear Energy. INL is the nation's center for nuclear energy research and development, and also performs research in each of DOE's strategic goal areas: energy, national security, science and the environment.*