

# Targets for Radioisotope Production, Neutron Scattering, Nuclear Data Measurements, and Exploring the Periodic Table

Kristian Myhre, Nathan Sims, Shelley Van Cleve,  
Susan Hogle, Jisue Moon

Isotope Applications Research Group  
Radioisotope Science and Technology Division  
Isotope Science and Engineering Directorate  
Oak Ridge National Laboratory

September 2022

# Bird's Eye View: Oak Ridge National Laboratory

Overview of ORNL activities related to nuclear target science and applications

*Stable isotope targets covered by Zach, Conner, & Foster talks*

Electrodeposition of Cf-251 targets for super heavy element studies



# Targets for Neutron Scattering at ORNL Spallation Neutron Source (SNS)

Ion Source produces 2.5 MeV H<sup>-</sup> ions

Linear Accelerator increases H<sup>-</sup> ion energy to 1 GeV

**Proton Accumulator Ring** bunches the H<sup>-</sup> ions and passes them through diamond stripper foil

**Spallation Target** circulates 20 tons of mercury during irradiation with the 1.4 MW 60 Hz proton beam

Future upgrades include power increase to 2.4 MW and a second target station



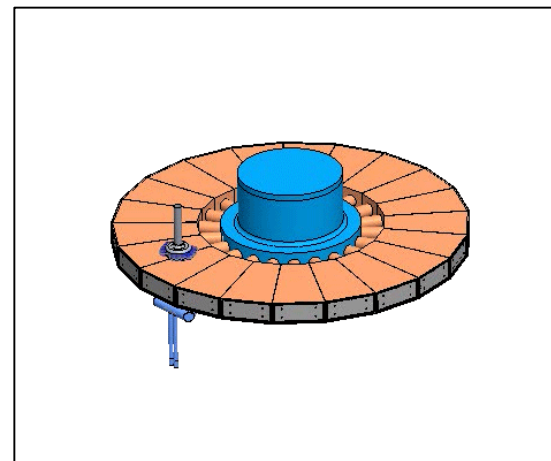
*Mercury target used to produce neutrons*

*~1  $\mu\text{m}$  thick diamond stripper foil used at SNS*

**[ornl.gov/facility/cnms](http://ornl.gov/facility/cnms)**



*Tungsten target concept for second target station*



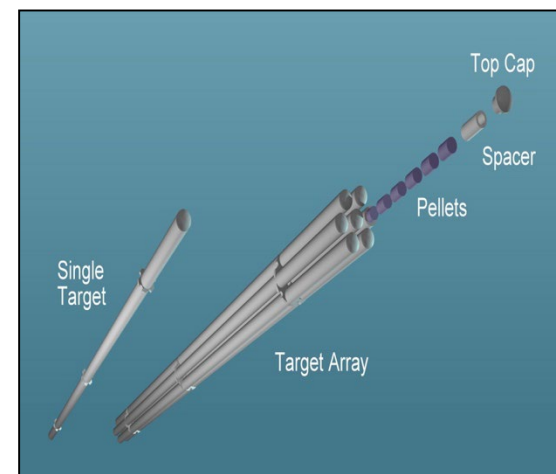
# Radioactive Targets for Isotope Production

ORNL produces a variety of targets with radioactive material for isotope production in nuclear reactors

Target materials include **Ra, Np, & Cm** for Ac, Pu, and Cf isotope production

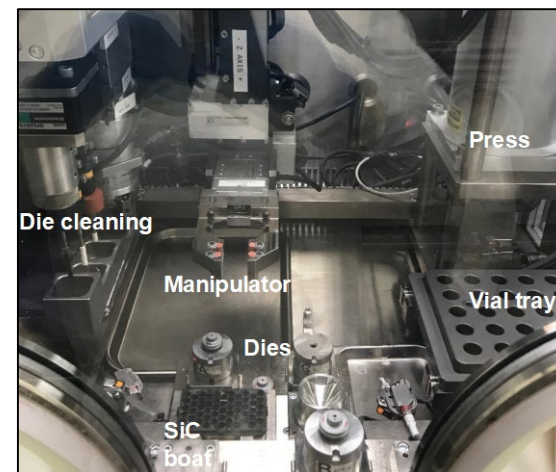
Automated and remote manufacturing are often required

Post-irradiation characterization facilities are growing (for actinides too!)



*Targets contain one or more pellets and often irradiated in arrays*

*Thousands of NpO<sub>2</sub>-Al pellets pressed per year with automated system in glovebox*



*Characterization techniques include nanoindentation, SEM, TEM, LECO, thermal desorption, and others*



# Actinide thin films for science and applications

ORNL primarily supplies raw radioisotope materials but can provide manufactured targets if needed

Isotopes have included Cf-251, Cf-252, and Am-241

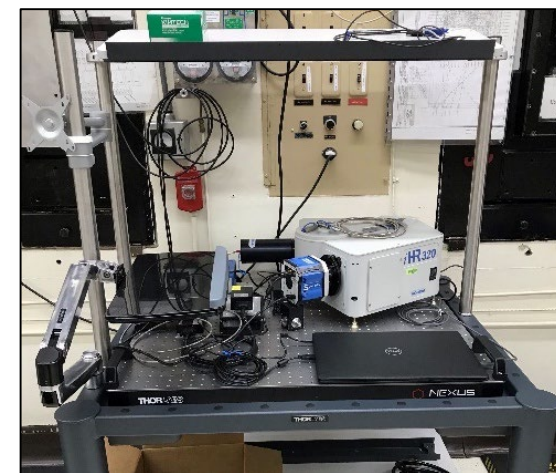
Recent R&D is focusing on real time thickness, mass, and spectroscopy

Goal is to enable real time control and deeper understanding of process



*>1 Ci of Cf-252  
electrodeposited for  
fission fragment source*

*Hot cell & glovebox  
Raman, absorption, &  
fluorescence systems*

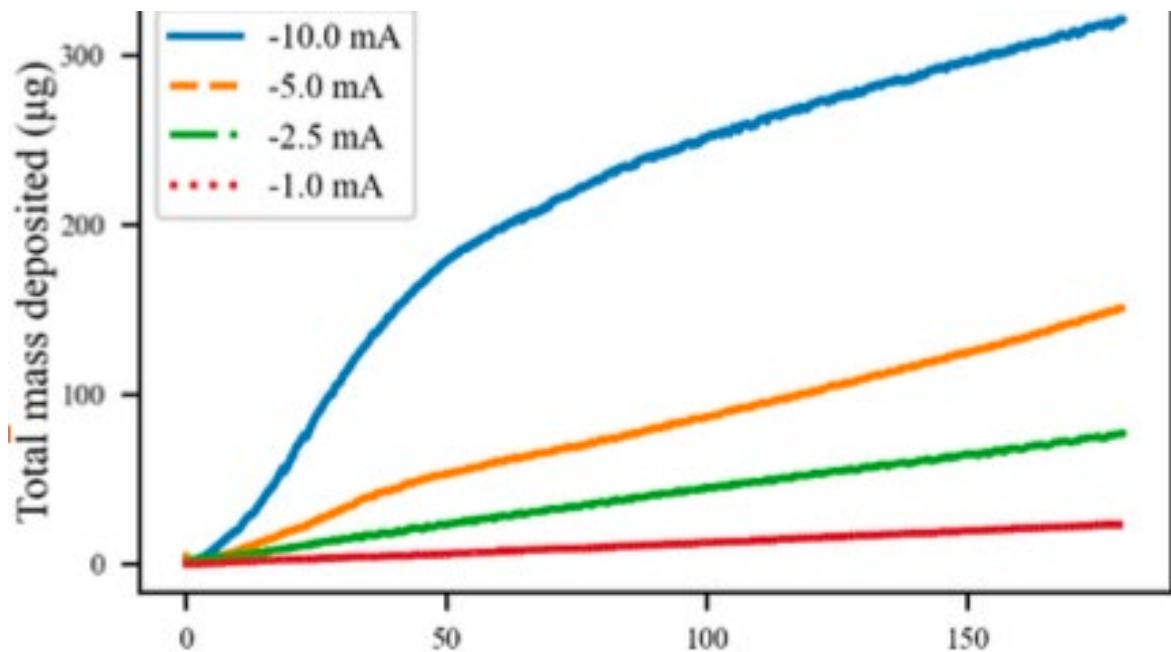


*Quartz Crystal  
Microbalance for real  
time mass data*



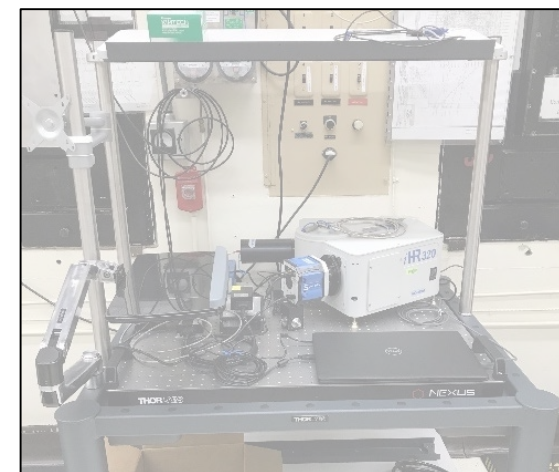
# Actinide thin films for science and applications

Example QCM data from La electrodepositions



Hot cell & glovebox  
Raman, absorption, & fluorescence systems

>1 Ci of Cf-252  
electrodeposited for  
fission fragment source



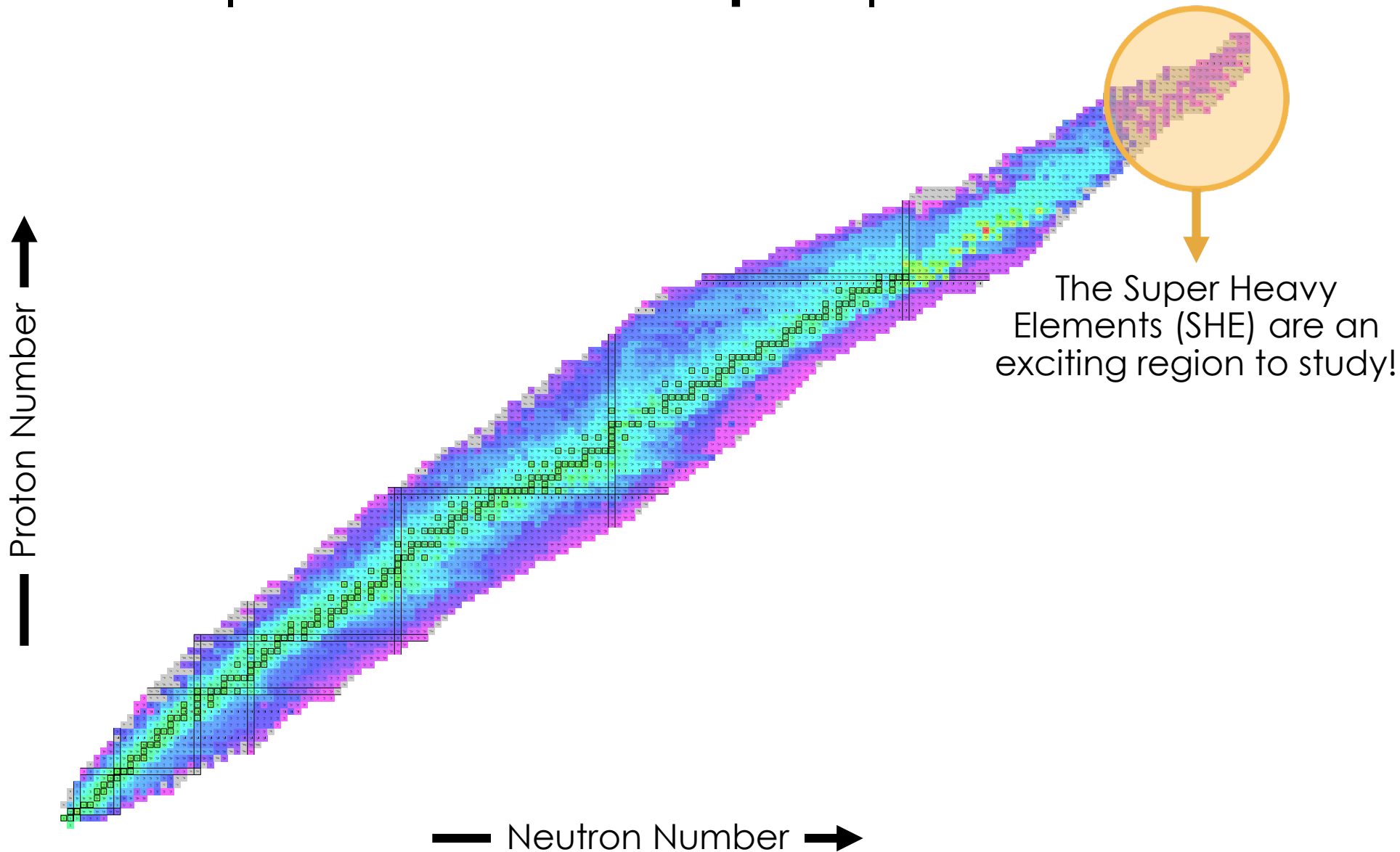
Quartz Crystal  
Microbalance for real  
time mass data



# Electrodeposition of Cf-251 targets for super heavy element studies



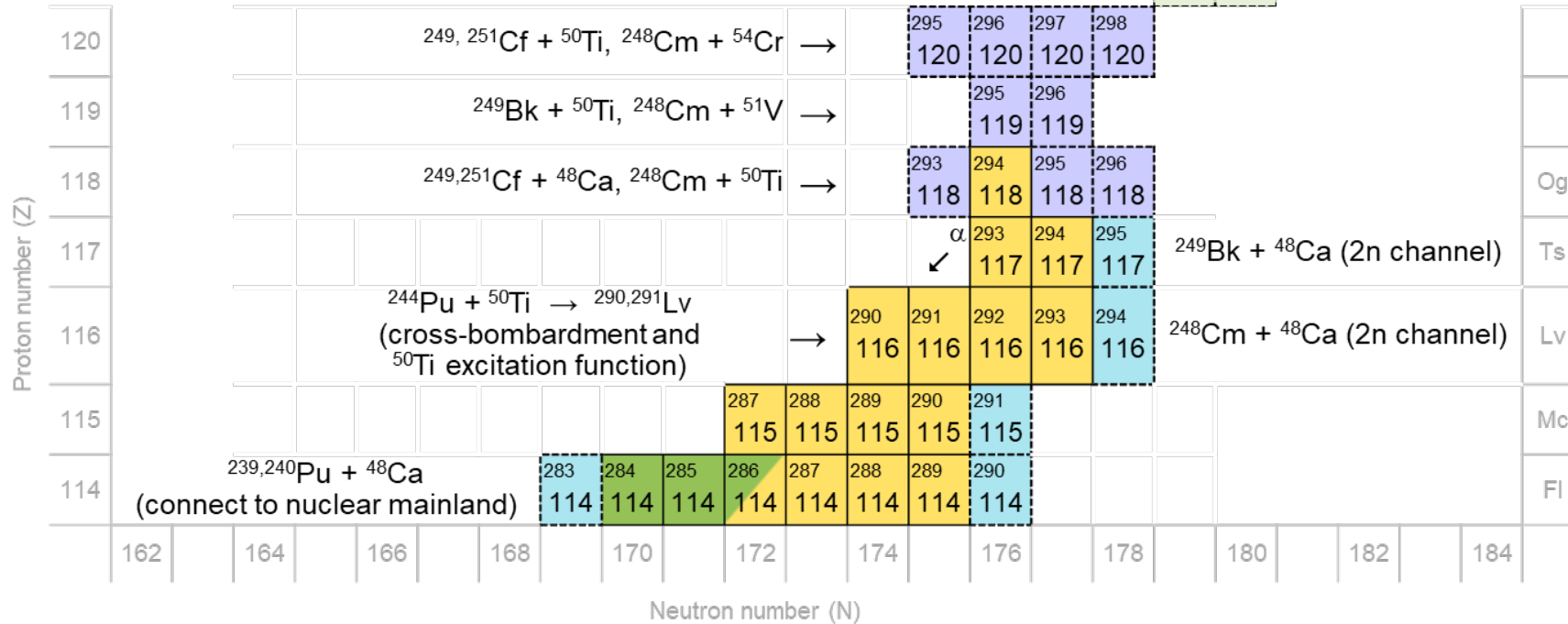
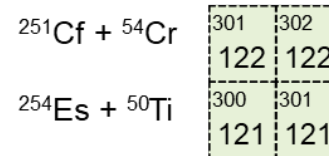
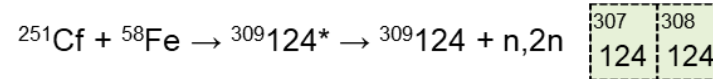
118 elements | 254 stable isotopes | >3000 radioisotopes





# Significant SHE research opportunities exist using actinide targets

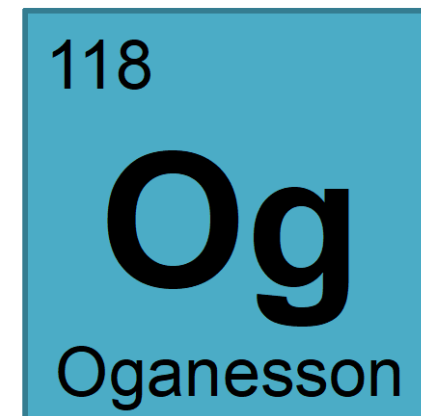
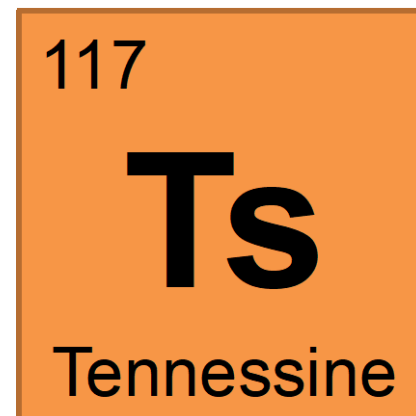
- New elements 119 and 120
- New heaviest isotopes of Og, Ts, Lv
- Light isotopes of Fl
- Excitation functions for  $^{50}\text{Ti}$
- Potential “cold fusion” path to N=184



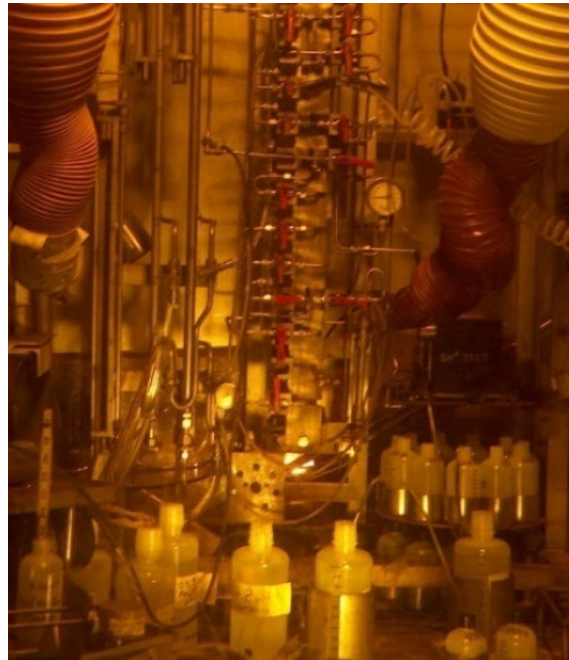
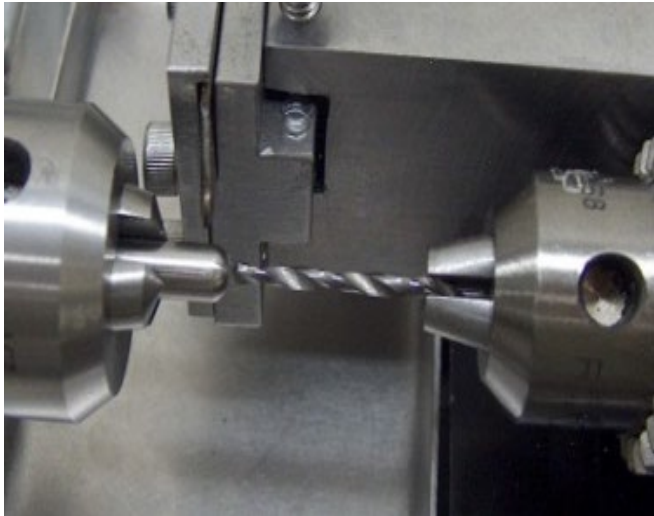
# Challenges of pushing beyond element 118

- Cross-sections are decreasing
- Beam intensities are increasing
- Heavier ions are needed
- Target materials are often limited

**All aspects of SHE research must  
be advanced to continue  
discovery science**



In 2014, rare mixed californium isotope material was recovered from decayed  $^{252}\text{Cf}$  sources at REDC



~16 mg of californium was recovered

---

### Target material isotopics

---

$^{249}\text{Cf}$	48.1%
-------------------	-------

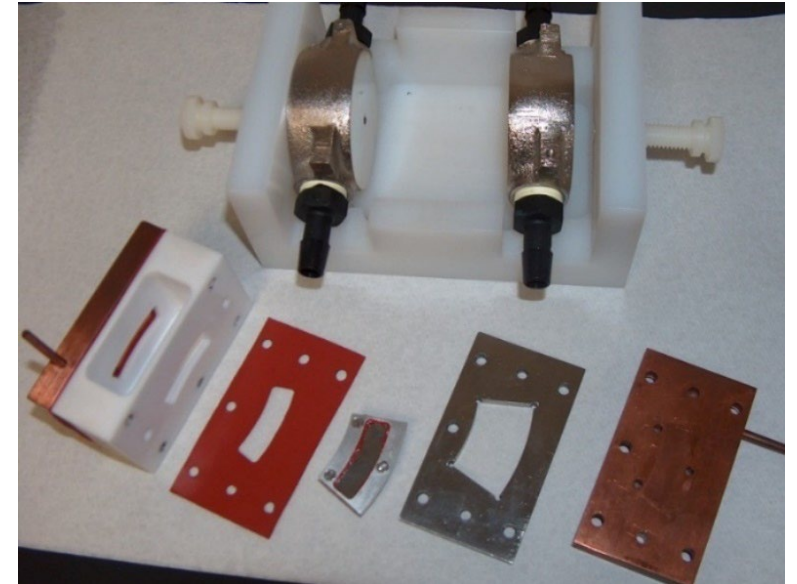
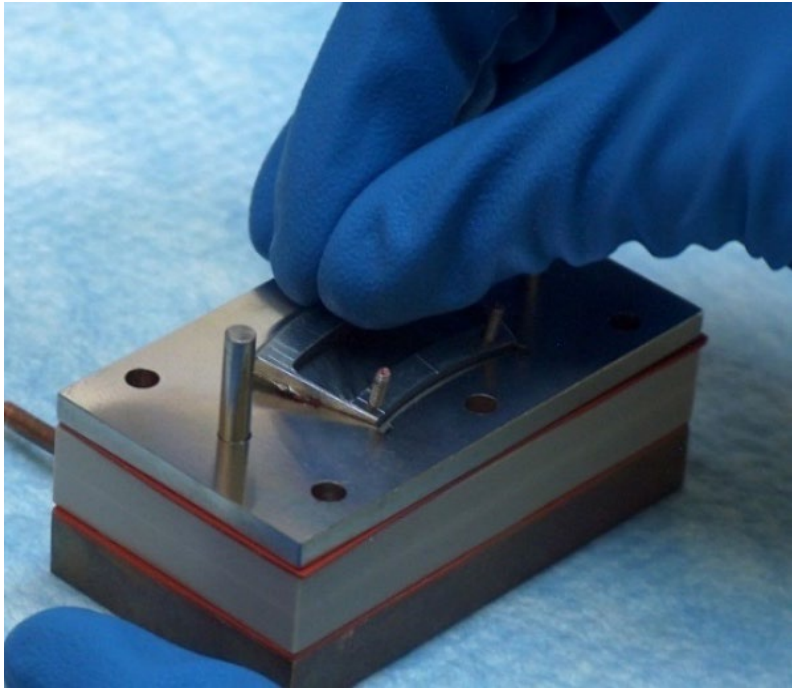
$^{250}\text{Cf}$	15.6%
-------------------	-------

$^{251}\text{Cf}$	36.3%
-------------------	-------

$^{252}\text{Cf}$	0.01%
-------------------	-------

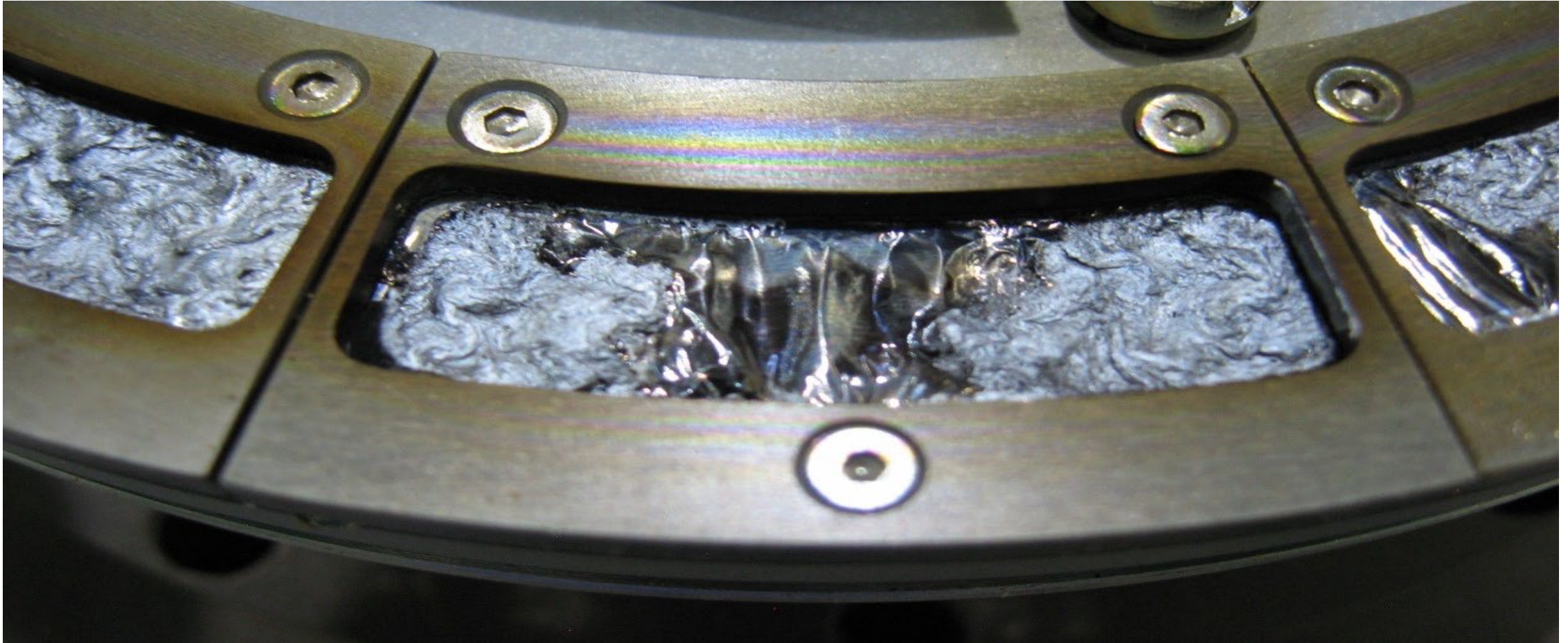
---

Twelve  $^{251}\text{Cf}$  enriched target segments were produced at ORNL using the isopropanol-isobutanol molecular plating technique

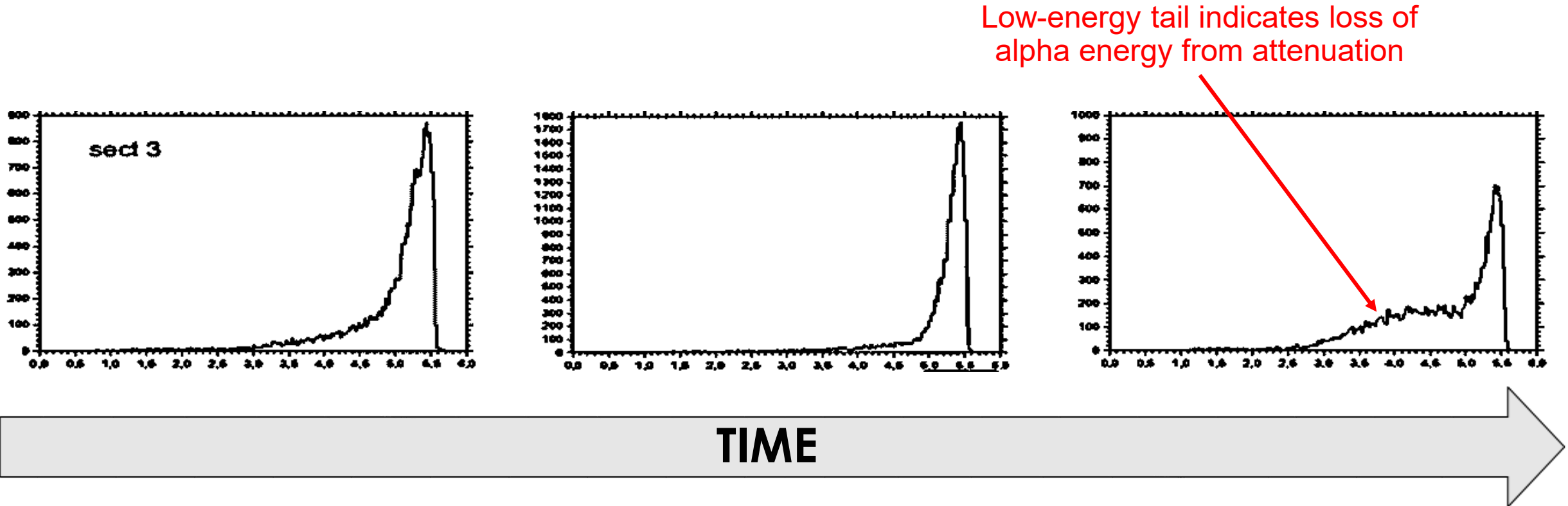


Deposition method adapted from **A. Vascon, et al., Nucl. Instr. And Meth. 655 (2011) 72**

An unknown film formed on the targets during irradiation at the Joint Institute for Nuclear Research (JINR)

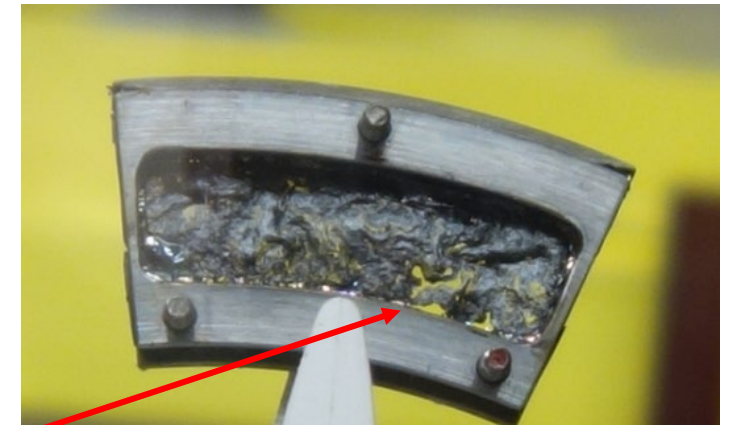
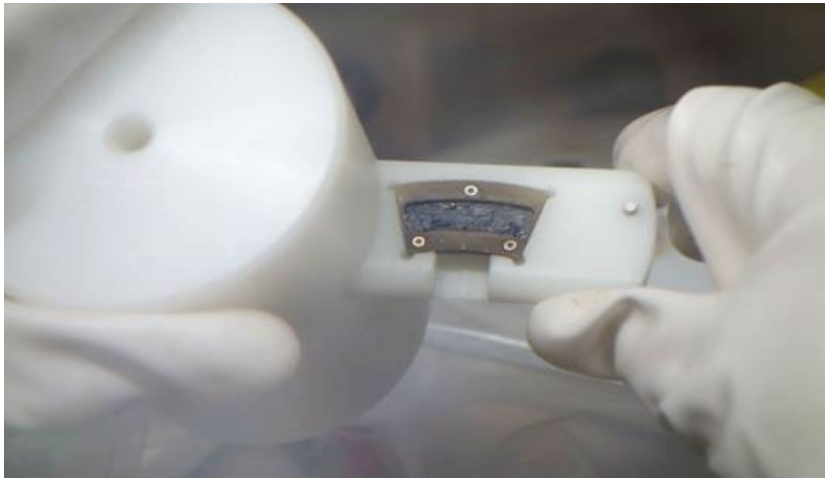


# Alpha spectroscopy showed the degrading quality of the targets during irradiation



Data provided from Vladimir Utyonkov at JINR

In 2016, the targets were shipped back to ORNL for recovery of the californium material and reproduction of the target segments for continued studies



Large areas of the foils were missing

# Target segment dissolution and analysis

Several 0.1 M  
HNO<sub>3</sub> rinses

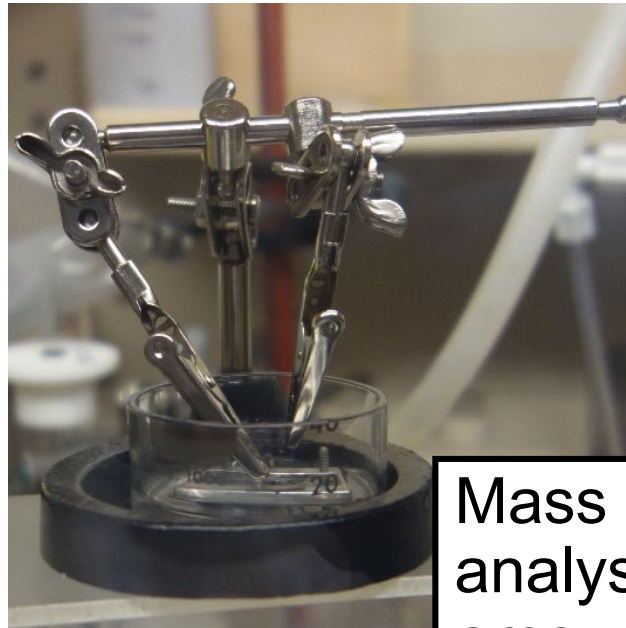
- Film mostly dissolved

Few drops of  
conc. HF

- Residual solids dissolved

Cation column  
purification

- 73% of original californium mass recovered



Mass spectrometry analysis showed large amounts of **silicon** in the solid films

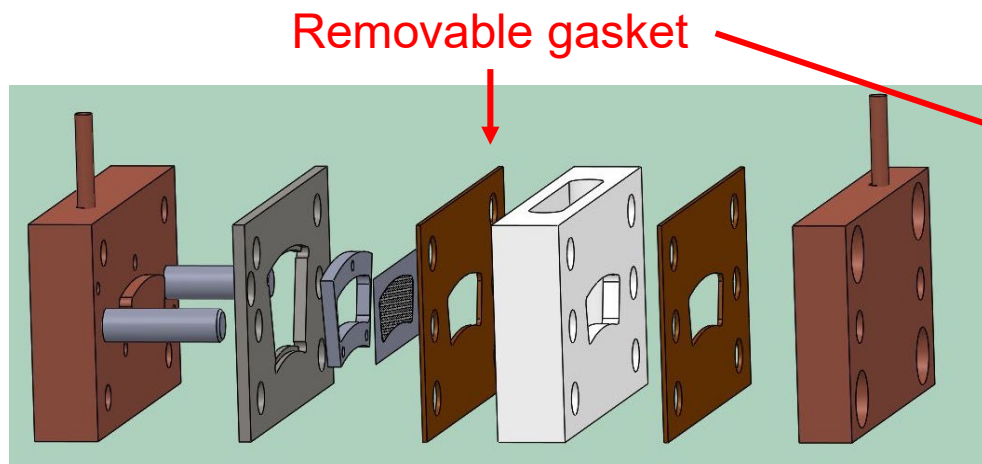
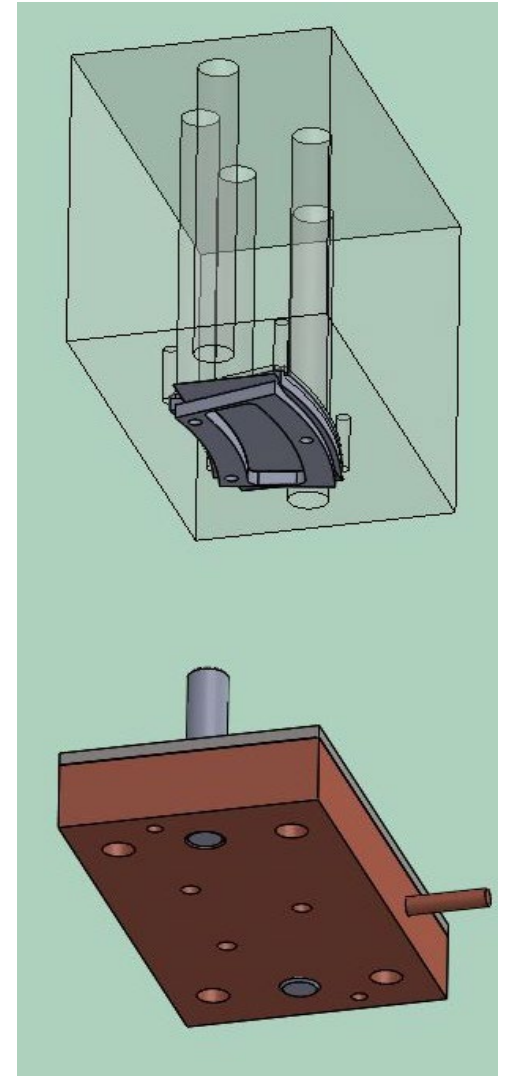




# New production method that results in an all-metal support structure for the actinide film was developed

## Electrodeposition redesign includes

- 1.5  $\mu\text{m}$  thick Ti foil is attached to unit with double-sided tape
- Graphite powder used to ensure the gasket does not stick to the titanium foil when removed
- Front frame section will be screwed onto disassembled portion and guided onto the titanium foil for assembly

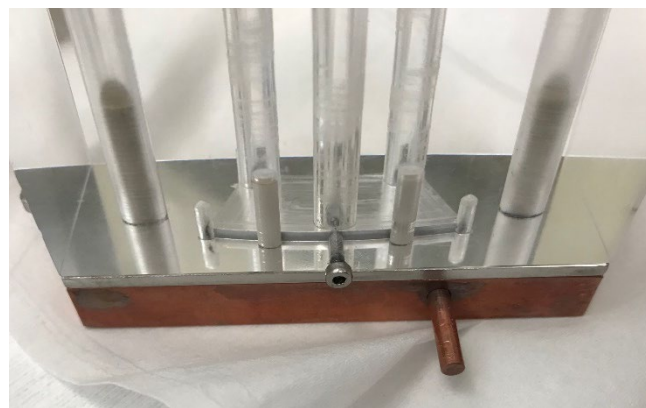


# New production methodology (continued)

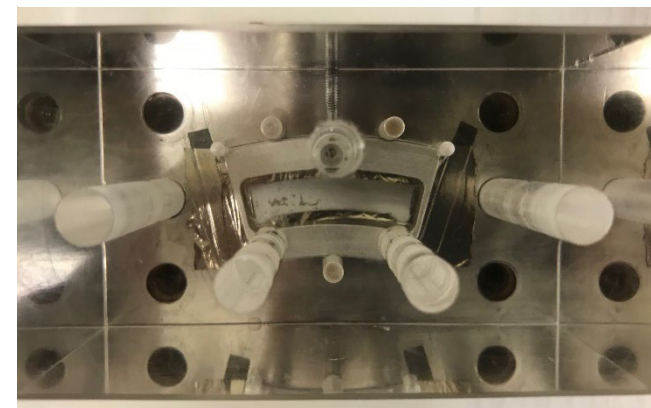
1. Deposition unit disassembled



2. Top frame holder lowered



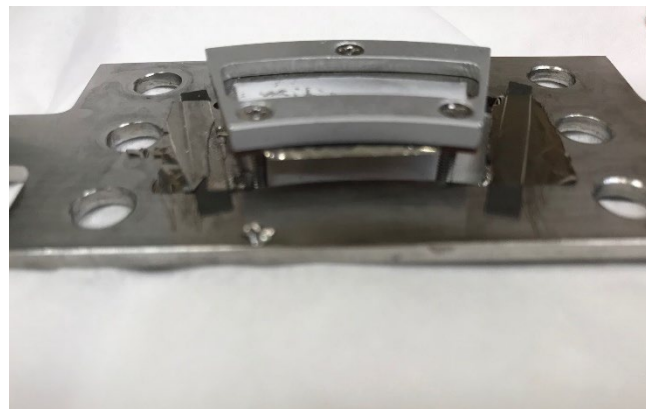
3. Frame screwed together



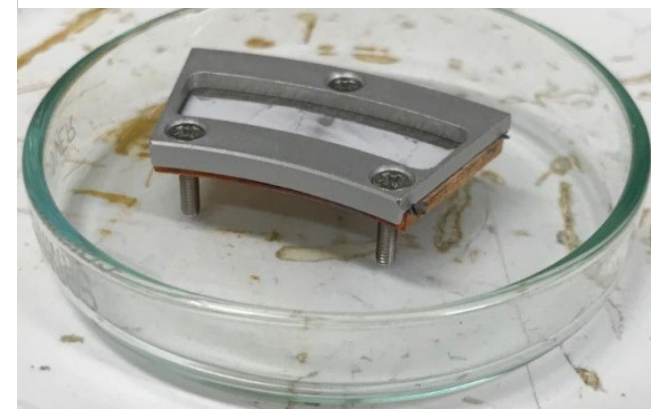
4. Sides of foil cut



5. Assembled target removed



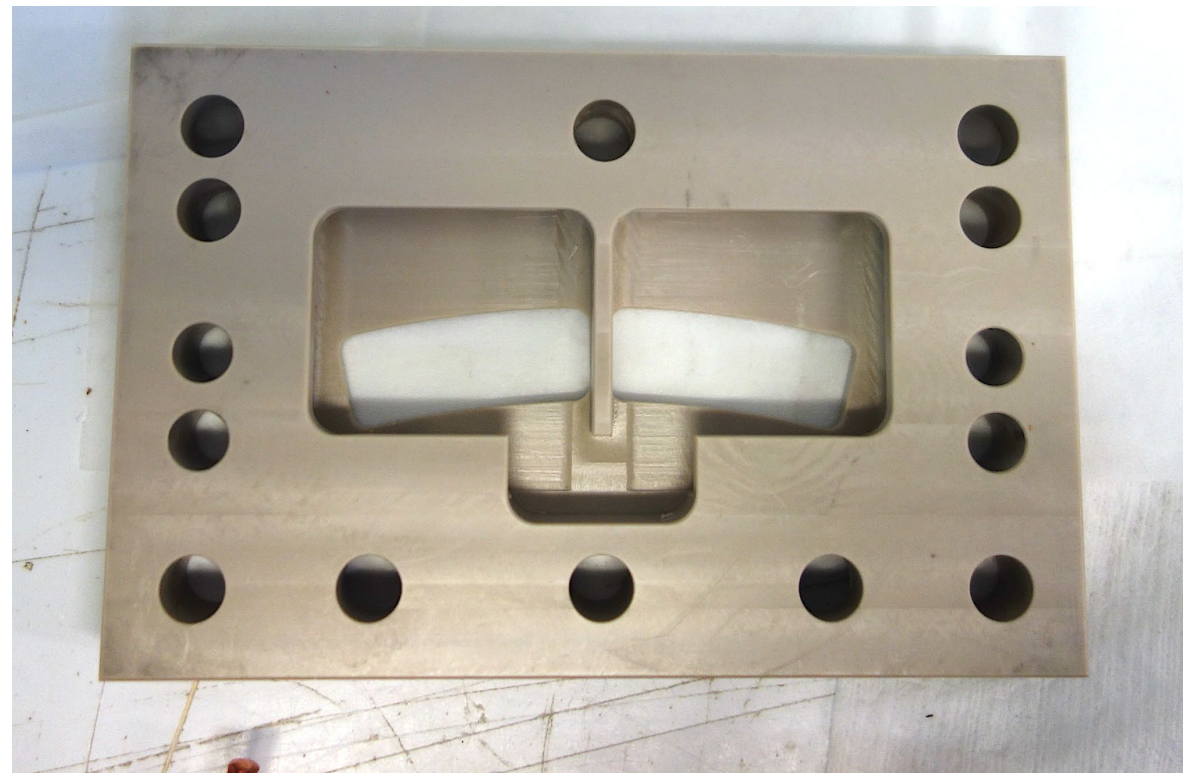
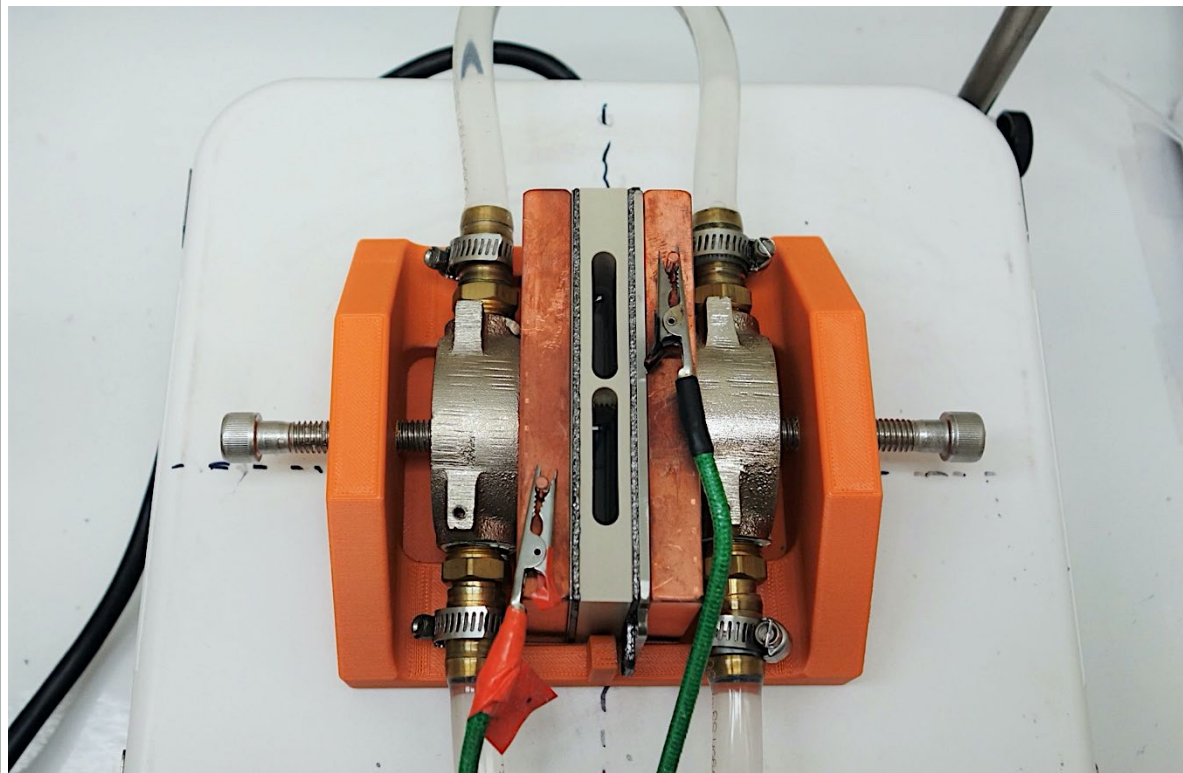
6. Target completed



Team decided double sector targets were needed, requiring further refinement of production methodology



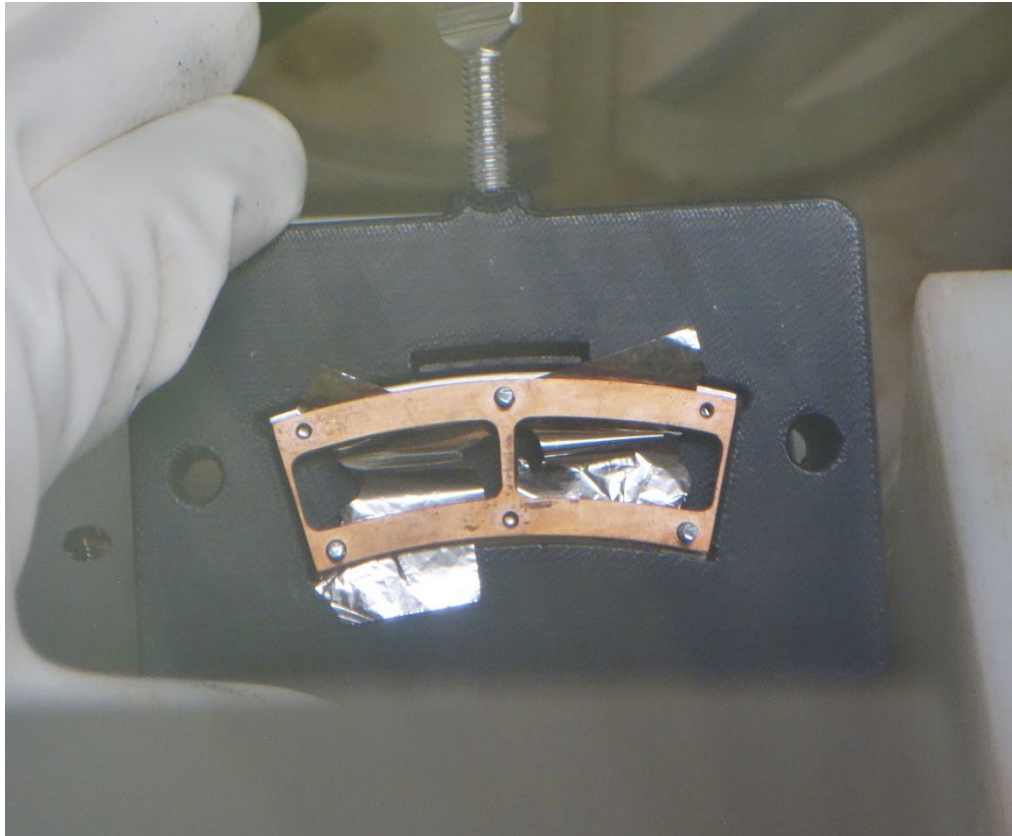
Redesign of the well was critical to ensure distinct deposition regions



After successful non-radioactive Sm tests on the bench and in the glovebox,  
**we were ready to produce the Cf targets!**

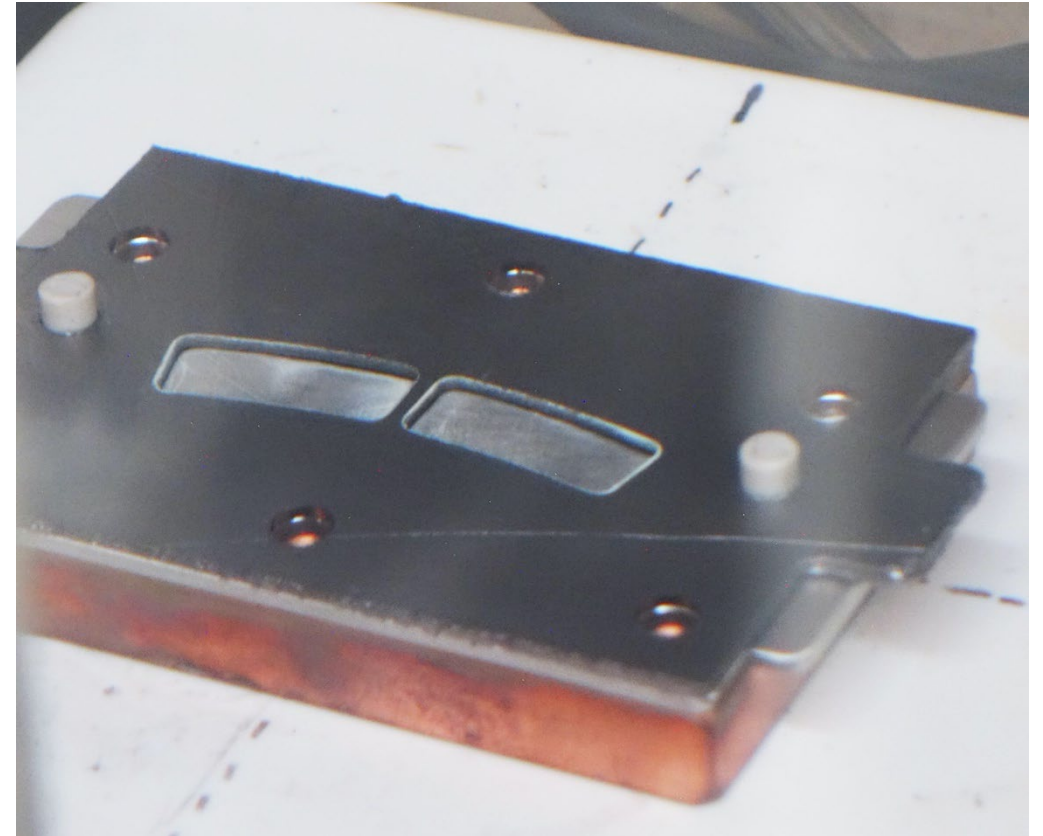
# But then... two disasters...

First Cf target broke during assembly



Frame holder mechanism found to be too fragile for repeated use

Second Cf target had low yield (22%)



Grey deposit was found on the rim of the graphite gasket

# Solution one: design a new target frame holder

Top of view of holder



Push guide



Bottom view of holder

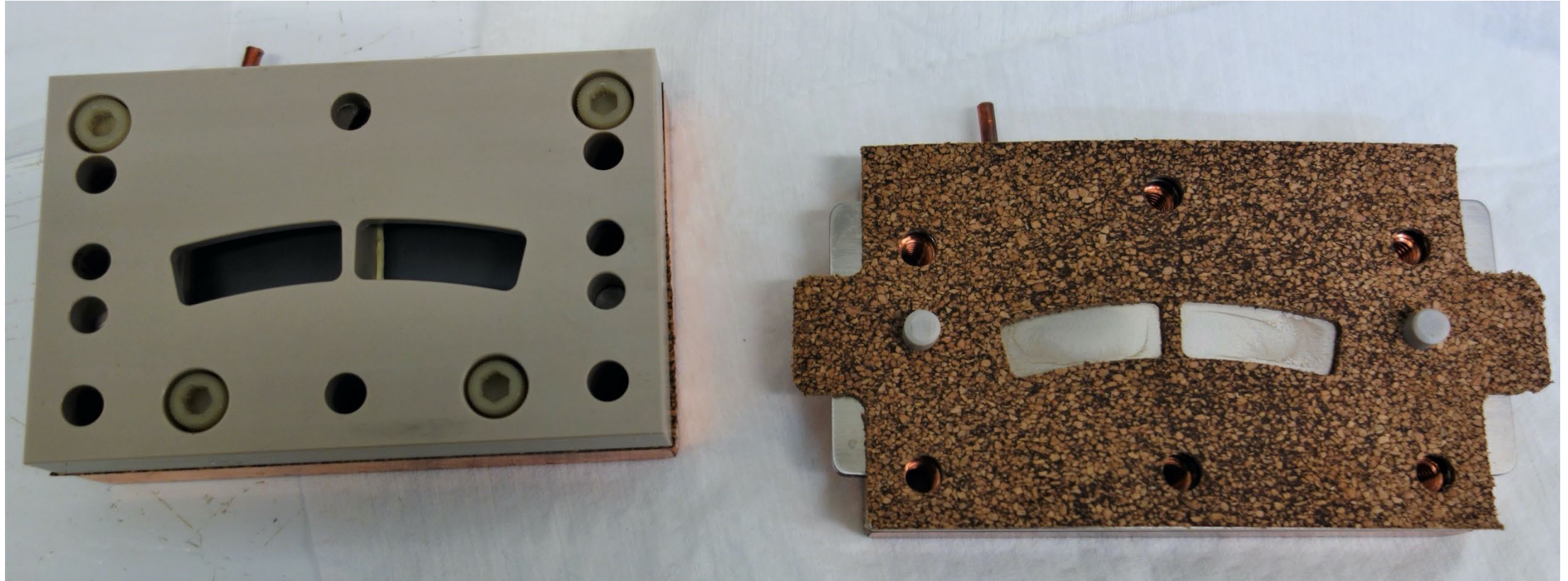


Holder with push guide inserted



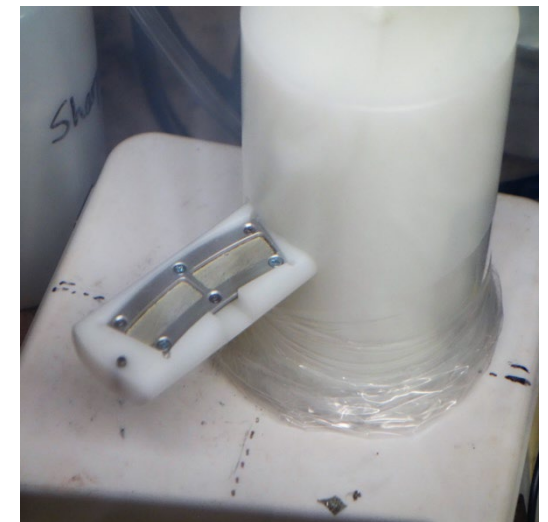
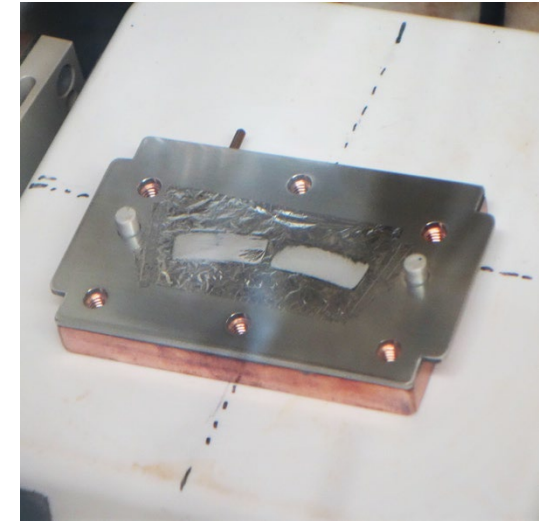
Small details were critical to success, including magnetic screwdriver, guides for screws, and push guide for target disengagement.

# Solution two: use a cork gasket



# Six Cf targets were successfully produced with an average deposition yield of 94%

Deposition Number	Percent Yield	Mass of Mixed Californium
1	Broken Target	n/a
2	22%	0.42 mg
3	97%	1.85 mg
4	98%	1.89 mg
5	90%	1.73 mg
6	94%	1.81 mg
7	88%	1.68 mg
8	95%	1.82 mg





# Summary

- ORNL is involved in a wide range of target production activities and eager to collaborate
- One of ORNL's roles in the Super Heavy Element community is to provide transuranic material and, in some cases, transuranic targets
- Six Cf-251 targets were produced at ORNL to help discover new nuclides of the heaviest elements
  - Production process has faced separation and fabrication challenges
  - Californium has been cleaned up successfully
  - Challenges to fabricating targets without a gasket in the final assembly were overcome
  - Average percent deposition was 94% for six consecutive targets

# Acknowledgments

Research sponsored by Department of Energy Office of Science Isotope Program and Office of Nuclear Physics

Shelley Van Cleve, Nathan Sims, Jisue Moon, Robert Sacci, Jonathan Morrison, Rose Boll, Julie Ezold, Clarice Phelps, Jordan Delashmitt, Ben Roach, Mike Zach, Dan Stracener, Jonathan Burns, James Roberto, Krzysztof Rykaczewski, Richard Mayes



ORNL Radiochemical Engineering and Development Center as well as Nuclear Materials Processing personnel



# Questions?

E-mail: [myhreckg@ornl.gov](mailto:myhreckg@ornl.gov)

