Recent Activities at Los Alamos National Laboratory Supporting Domestic Production of ^{99}Mo

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2016 ^{99}Mo Topical Meeting

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LANL Support for Domestic ^{99}Mo Production

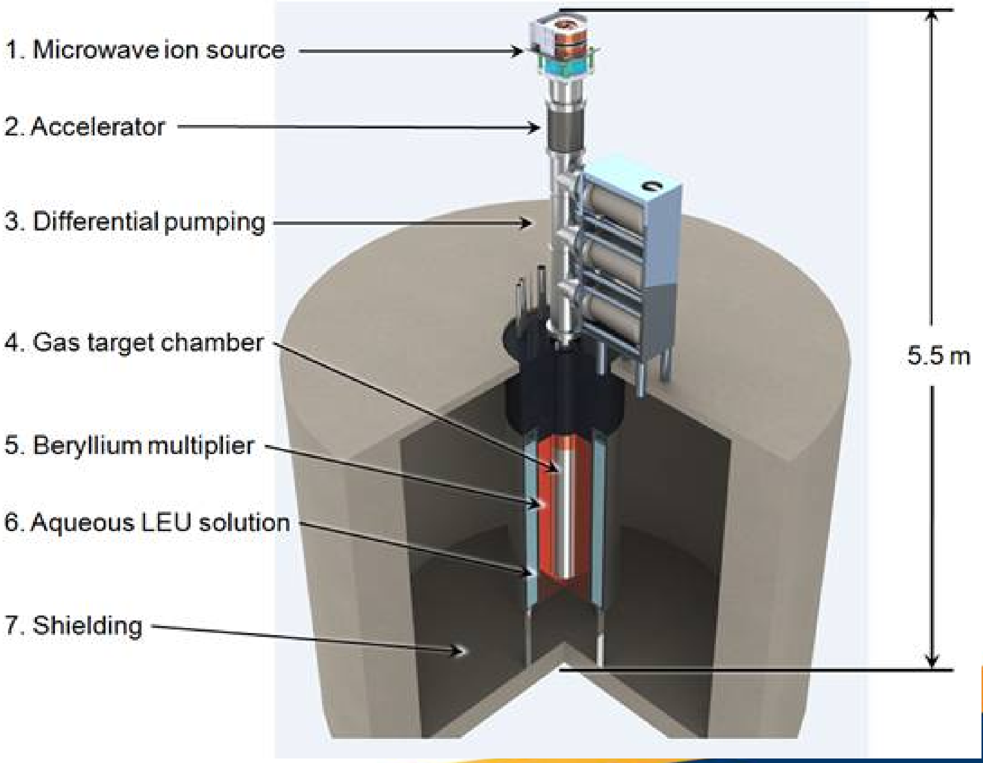
- As part of the NNSA Material Management and Minimization (M^3) Program, LANL is supporting:
  - NorthStar Medical Radioisotopes with the electron accelerator production of ^{99}Mo from ^{100}Mo(γ,n)^{99}Mo.
  - Shine Medical Technologies with the production of fission product ^{99}Mo from a DT accelerator driven subcritical uranium salt solution.

SHINE Medical Technologies Production Overview

SHINE Medical Technologies will produce fission product ^{99}Mo in a subcritical accelerator driven low enriched uranium salt solution

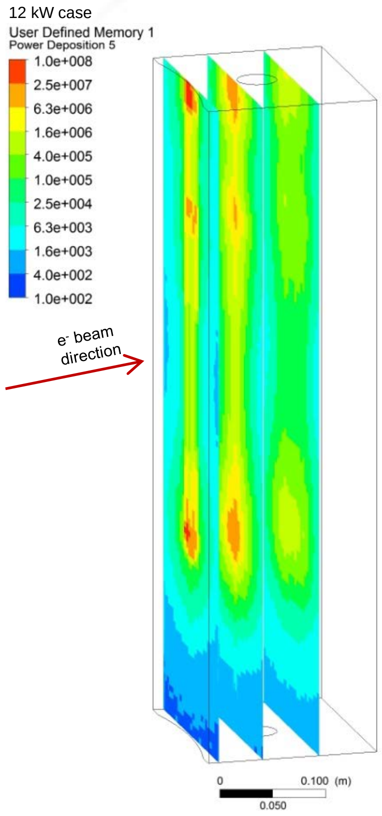
LANL Support Areas:

- Thermal Hydraulics and Coupled Neutronics Modeling
- Spectroscopic Analysis Technique for Uranium Concentration Measurement in Solutions

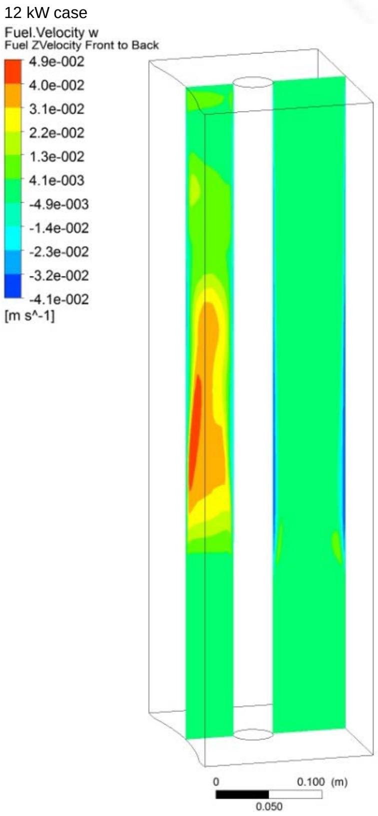


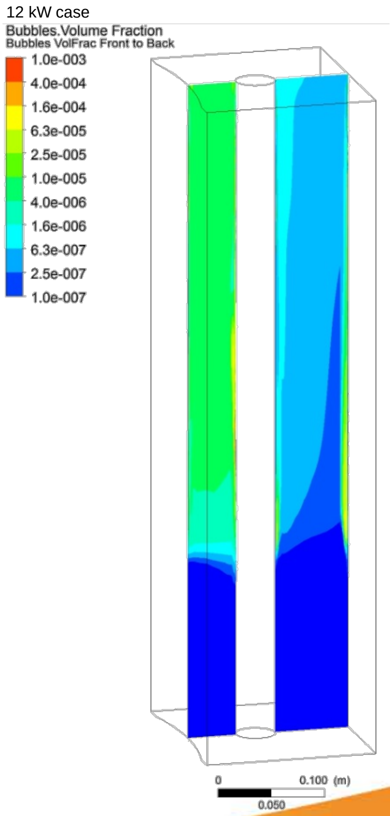
Thermal-Hydraulics Modeling to Support SHINE

- Two computational fluid dynamics (CFD) studies performed using Fluent
  - Heat transfer by natural convection enhanced by bubble generation
  - Multiphase models with liquid-bubble interaction



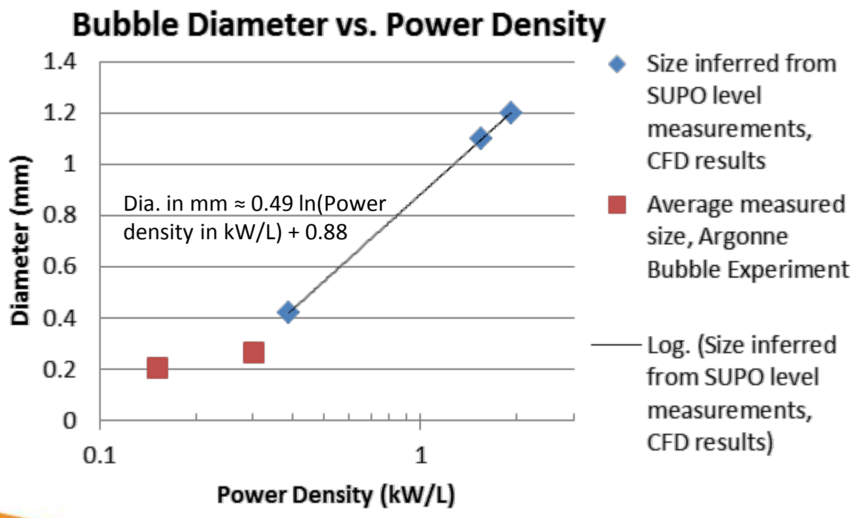
- Computational Study of Argonne Bubble Experiment
  - 35 MeV electron beam rastered on uranyl sulfate, generating heat and radiolytic gas bubbles.
  - 12 kW irradiation produced ~ 0.3 kW/L.
  - MCNP model computes power deposition profile from beam intensity measurement.
  - 3-D CFD model predicts steady state liquid temperatures and bubble volume fractions.

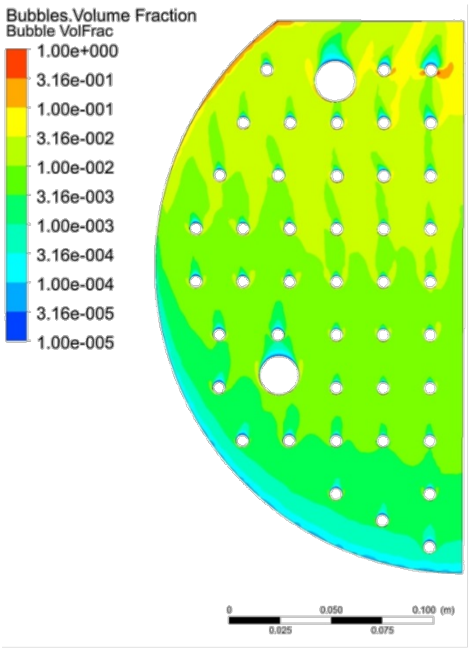


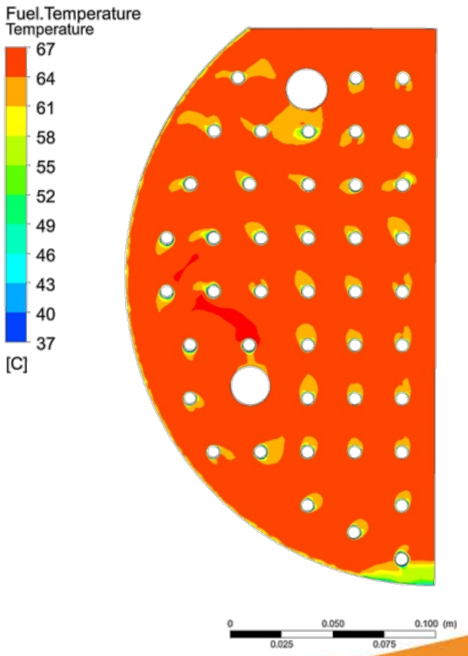


Thermal-Hydraulics Modeling to Support SHINE

- "Super-Power" reactor (SUPO) radiolytic bubble size study
  - SUPO operated at LANL 1951-1974 at power levels of 3-40 kW (0.2 - 3.0 kW/L).
  - 2-D axisymmetric model predicts steady-state temperatures, volume fractions
    - Gaussian power deposition profile calculated from neutron flux measured in "glory hole".
    - Gas generation specified using reported radiolytic gas generation rate: 2.78e-9 kg/s/W. (G = 1.5)
    - Bubble size determined by matching CFD volume fraction results to volume-fractions inferred from level height measurements for 5.1, 20.1, and 25.2 kW cases.







Thermal-Hydraulics Modeling to Support SHINE

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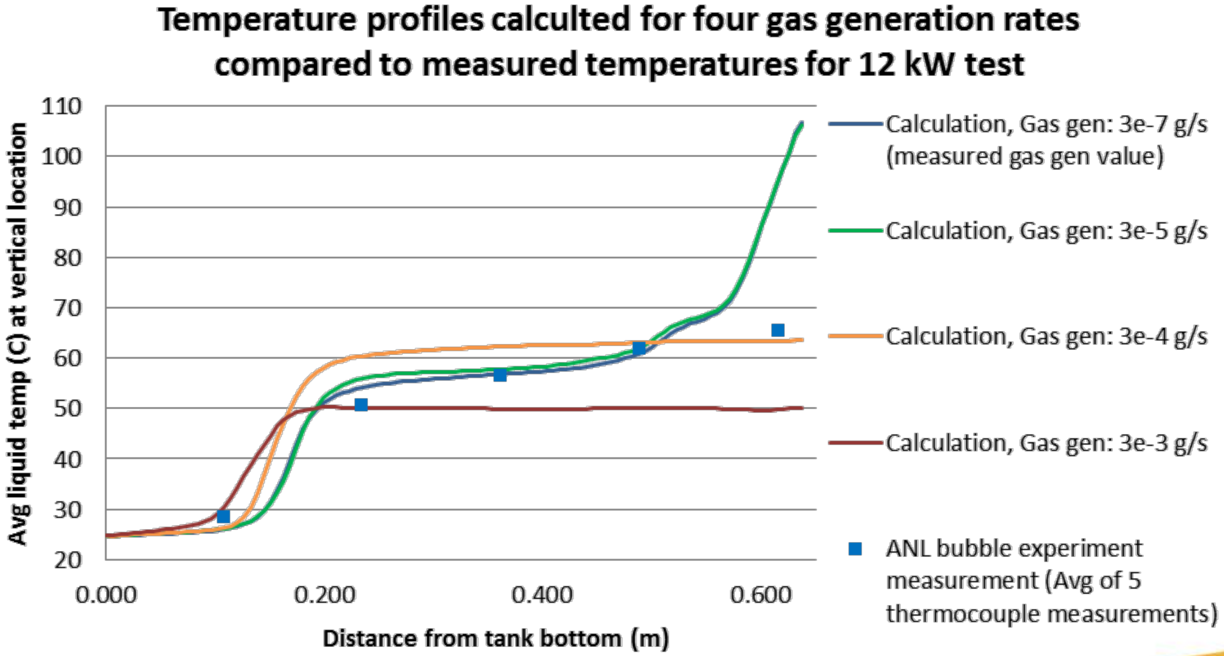
- A gas generation sensitivity analysis has been performed for the Argonne Bubble Experiment CFD model
  - Temperatures calculated using measured gas generation rates match measurements reasonably well, except near the top of the tank.
  - Uncertainty in gas generation rate measurement is the most likely reason for the observed temperature difference.



Uranium Solutions

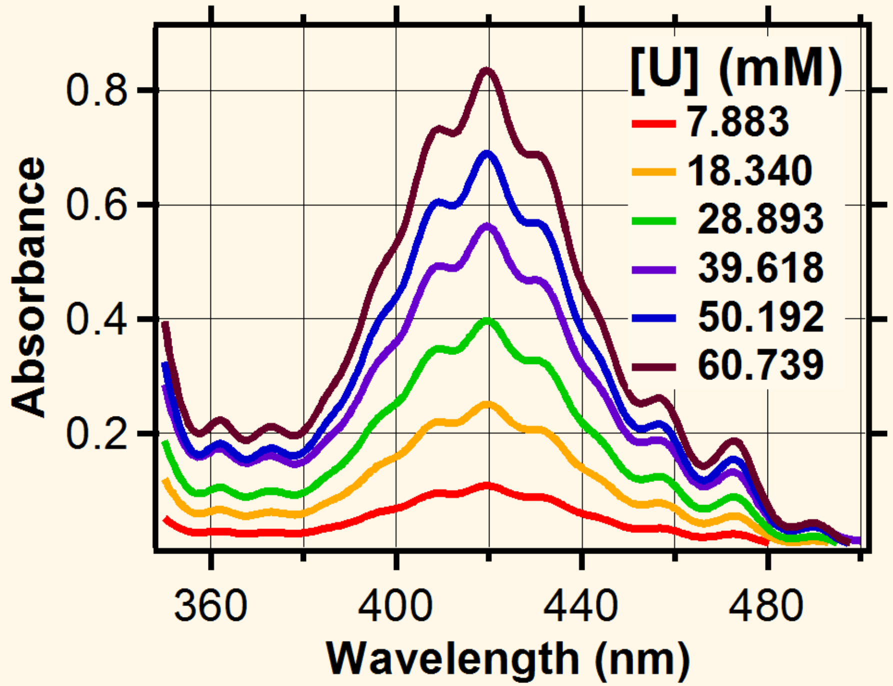
A rapid and robust method for uranium analysis in solution is required to meet US NRC regulations.

Left to right.

- ***Uranium metal standard dissolved in HNO_3,***
- ***A sample of this solution converted to solid $UO_3 \cdot xH_2O$, and***
- ***Resulting solid material dissolved in 1 M H_2SO_4 to yield a standard solution***

Spectroscopic Analysis Technique for Uranium Concentration Measurement in Solutions

A spectroscopic analysis technique has been developed for accurately measuring uranium concentration in solutions



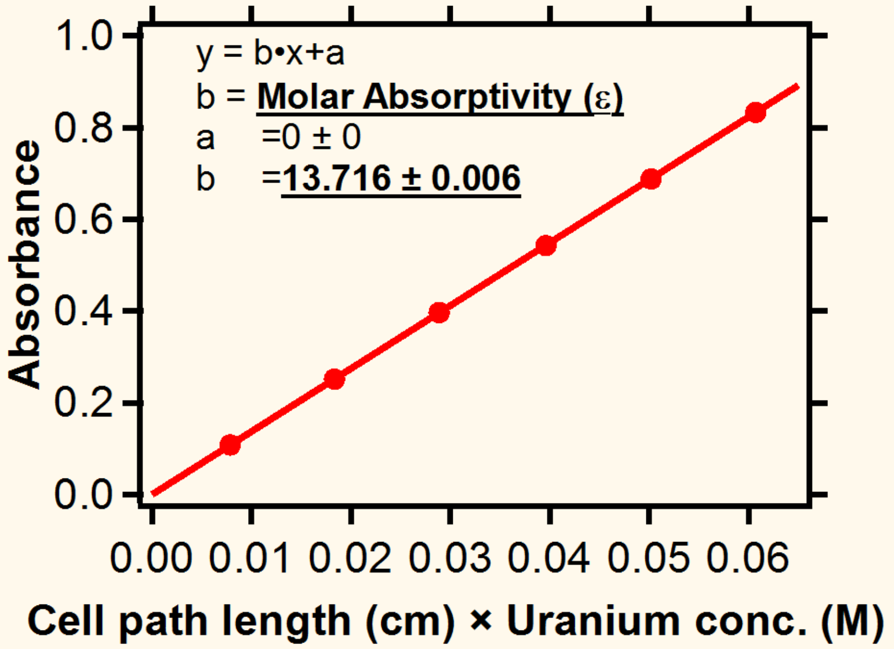
Uranium Standard Solution Spectra in 1 Molar Sulfuric acid

Beer-Lambert Law

$$A = \varepsilon . c . l$$

Where A = absorbance, ε = molar absorptivity, c = concentration (Molar) and l = spectroscopy cell path length (typically 1 cm)

Results and Equipment



Molar absorptivity for one Starna Cell at 22.5 °C

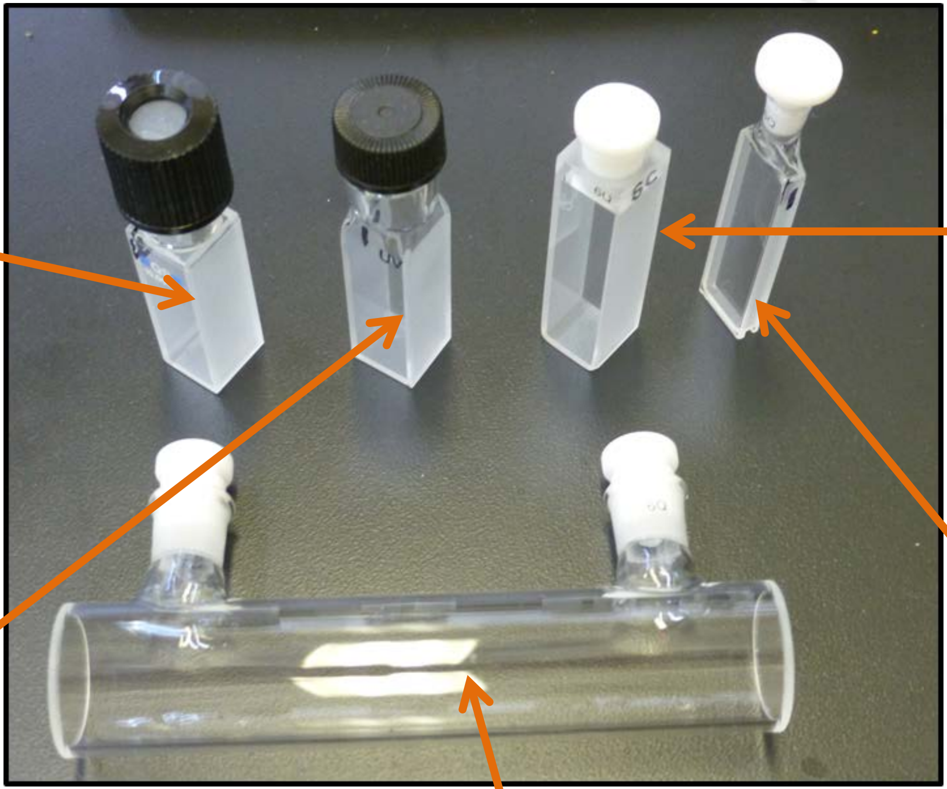
Agilent Technologies Cary 60 spectrophotometer with Peltier temperature control unit

Mettler Toledo DM50 Density Meter.

LA-UR-16-21310

Cell Manufacturer and Path Length

Hellma Analytics
(10.00 ± 0.01 mm)



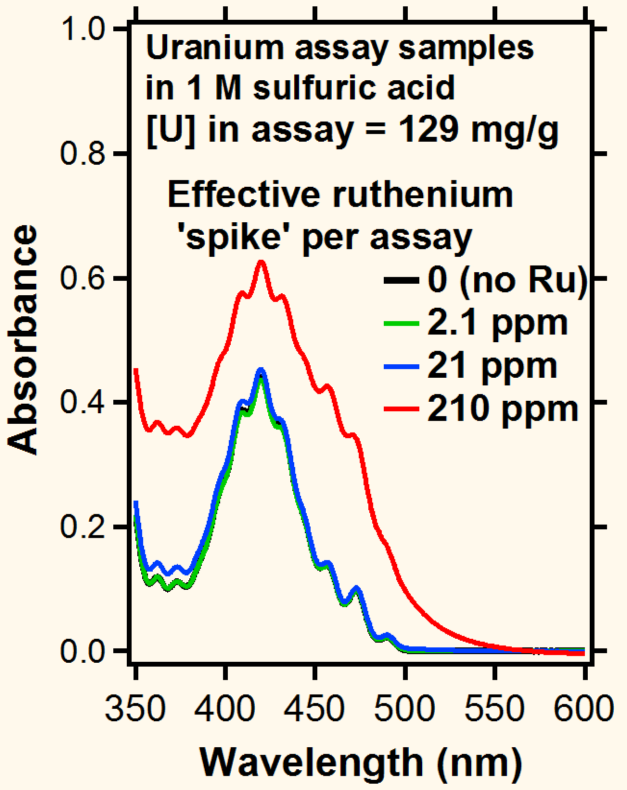
Starna Cells
(10.00 ± 0.01 mm)

NSG Precision
(10.00 ± 0.02 mm)

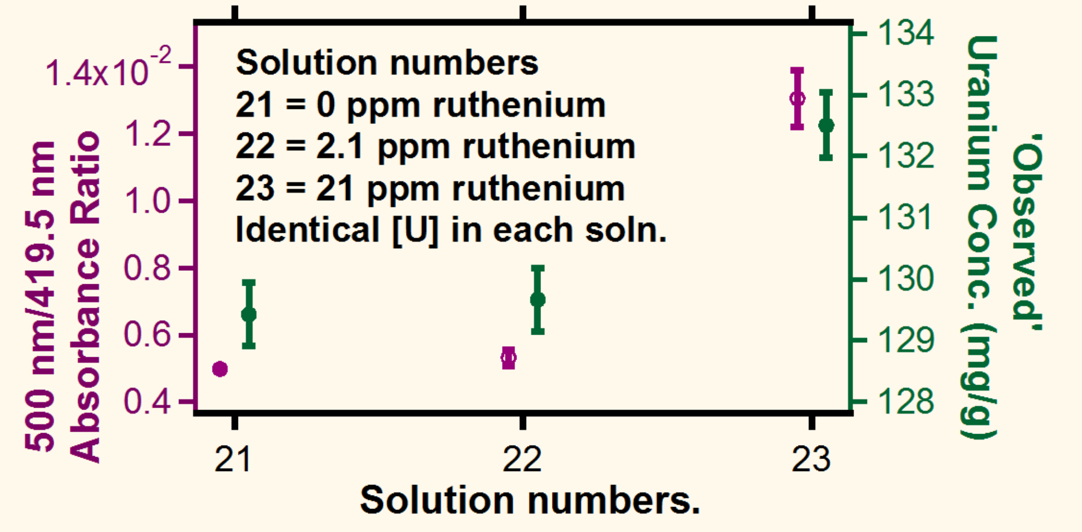
Starna Cells
(2.00 ± 0.01 mm)

Starna Cells
(100.00 ± 0.02 mm)

Analysis with the Presence of Impurities (Fission Products)

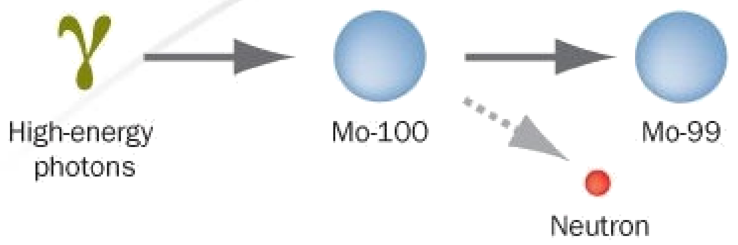


Colored Contaminant Impact

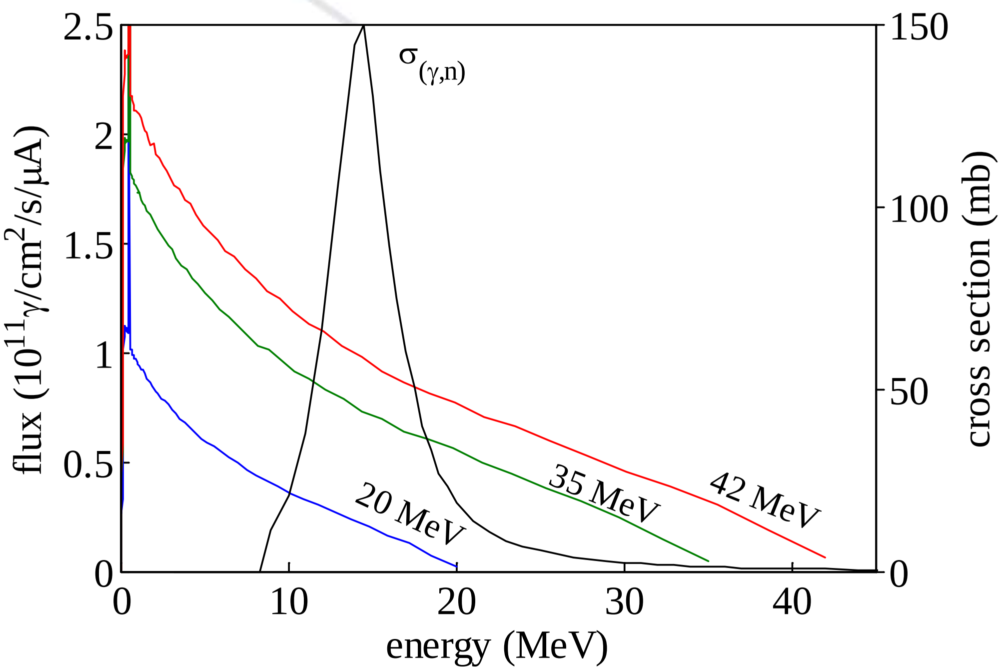


Change in absorbance ratios, and associated errors, can be used to 'invalidate' contaminated samples

NorthStar Electron Accelerator Production



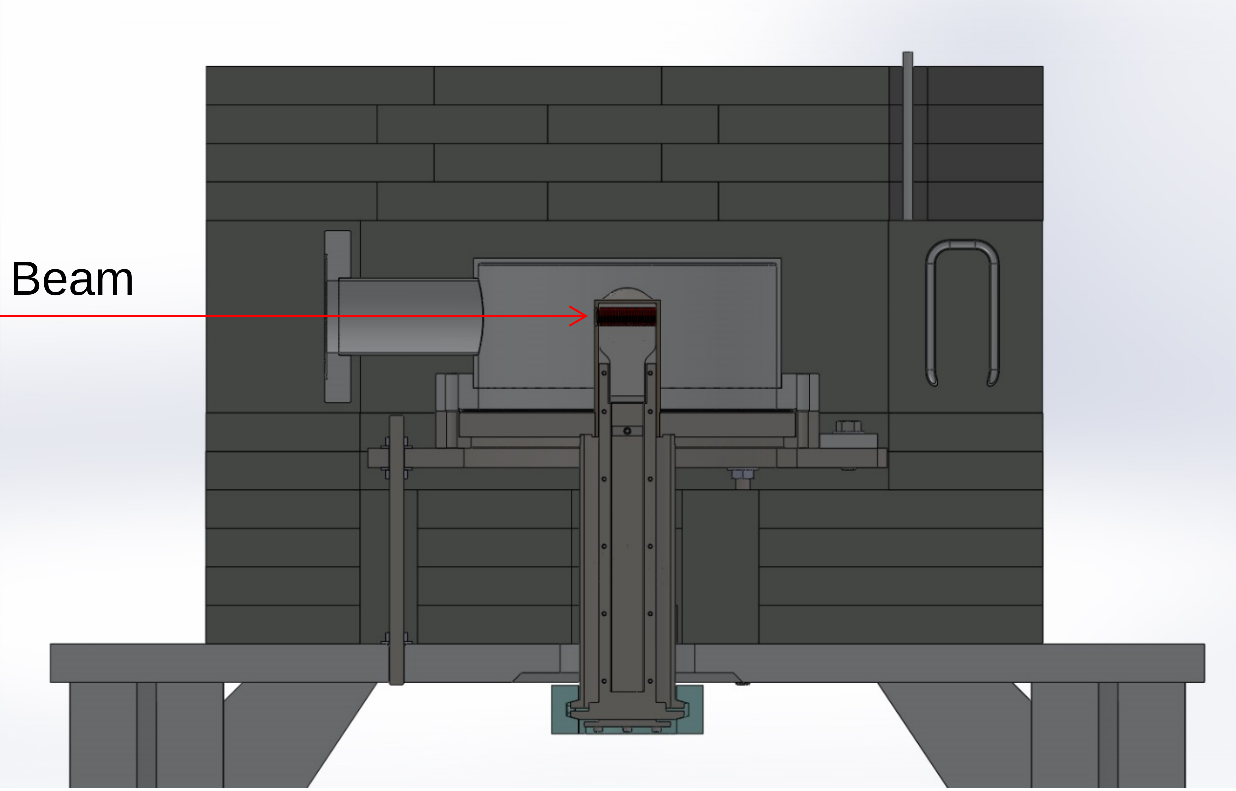
- The NorthStar process uses an electron accelerator to create a high flux of bremsstrahlung photons in enriched ^{100}Mo targets to create ^{99}Mo through the photonuclear reaction $^{100}Mo(\gamma,n)^{99}Mo$.
  - Reaction threshold is 9 MeV.
  - Peak cross section is 150 mb at 14.5 MeV.
- We are exploring electron beams in the 35-42 MeV range.



Average bremsstrahlung photon spectra produced with 20, 35, and 42 MeV electron beams in a Mo target compared to the photonuclear cross section of ^{100}Mo.

LANL 12 mm Diameter Mo Target for Testing at ANL

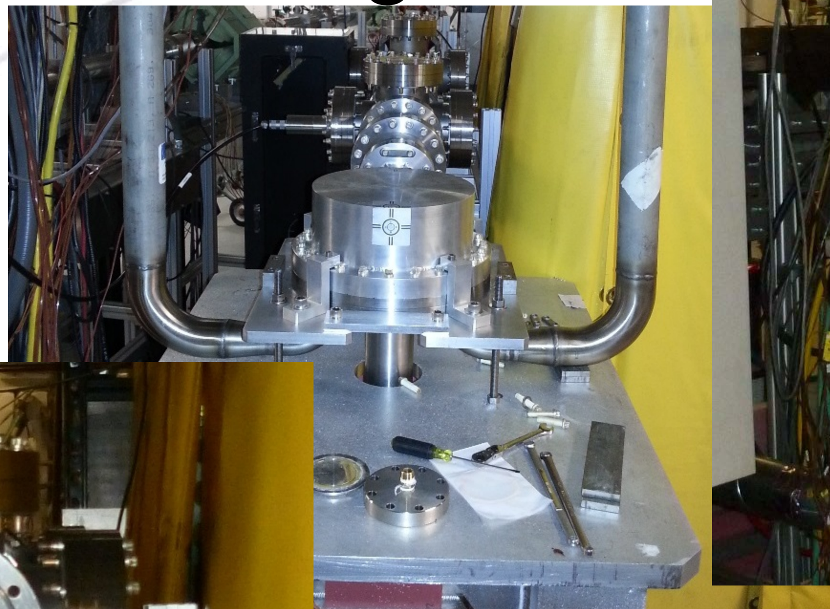
Target Side View

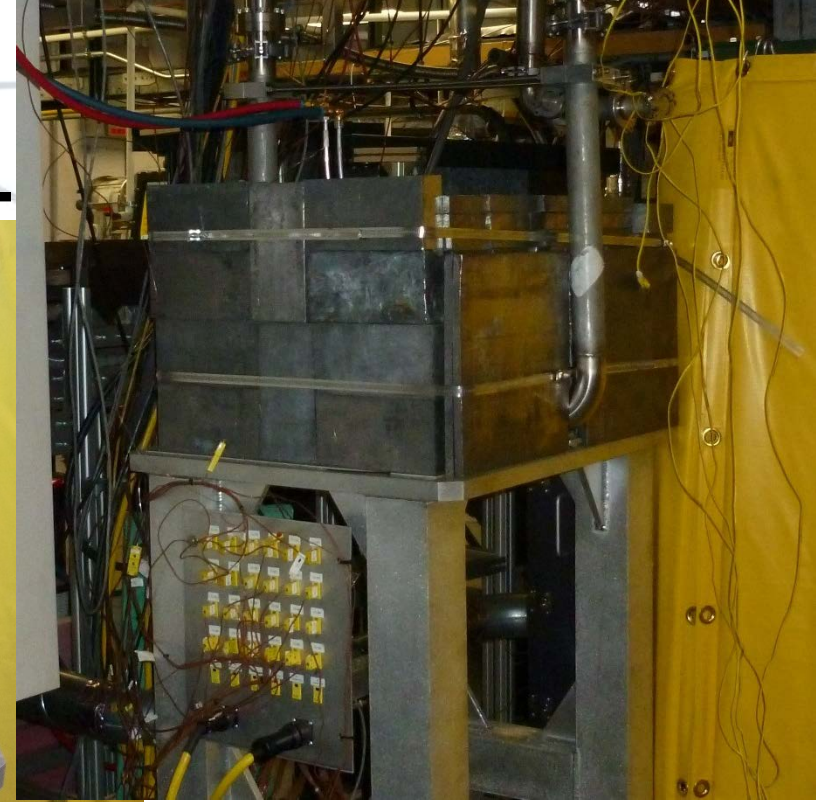


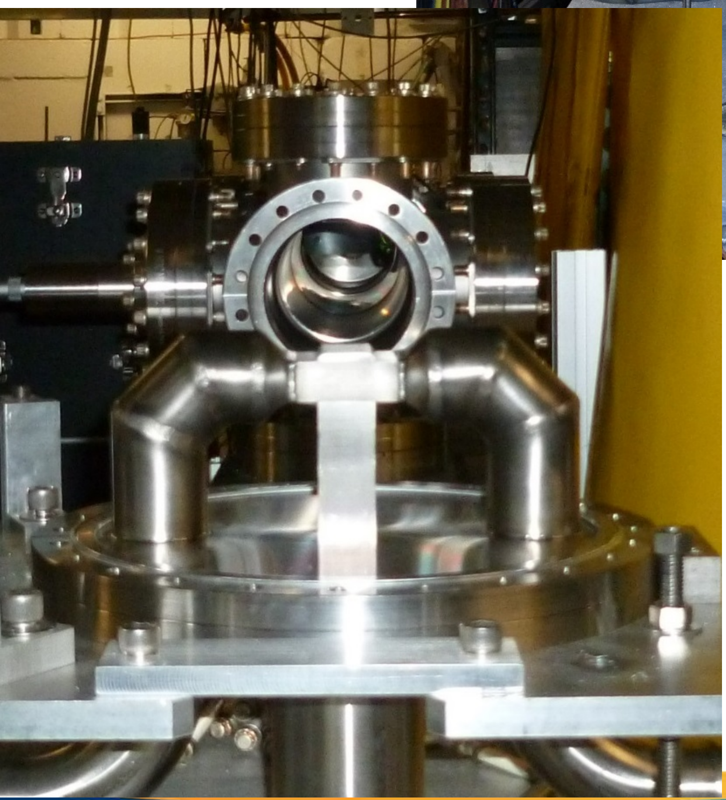
LANL designed single sided target for thermal and production tests

Target consisting of 25, 12 mm diameter, 1 mm thick disks with 1 mm cooling gaps

12 mm Diameter Target Installed for Testing at ANL







Experiments completed with this target in the last year:

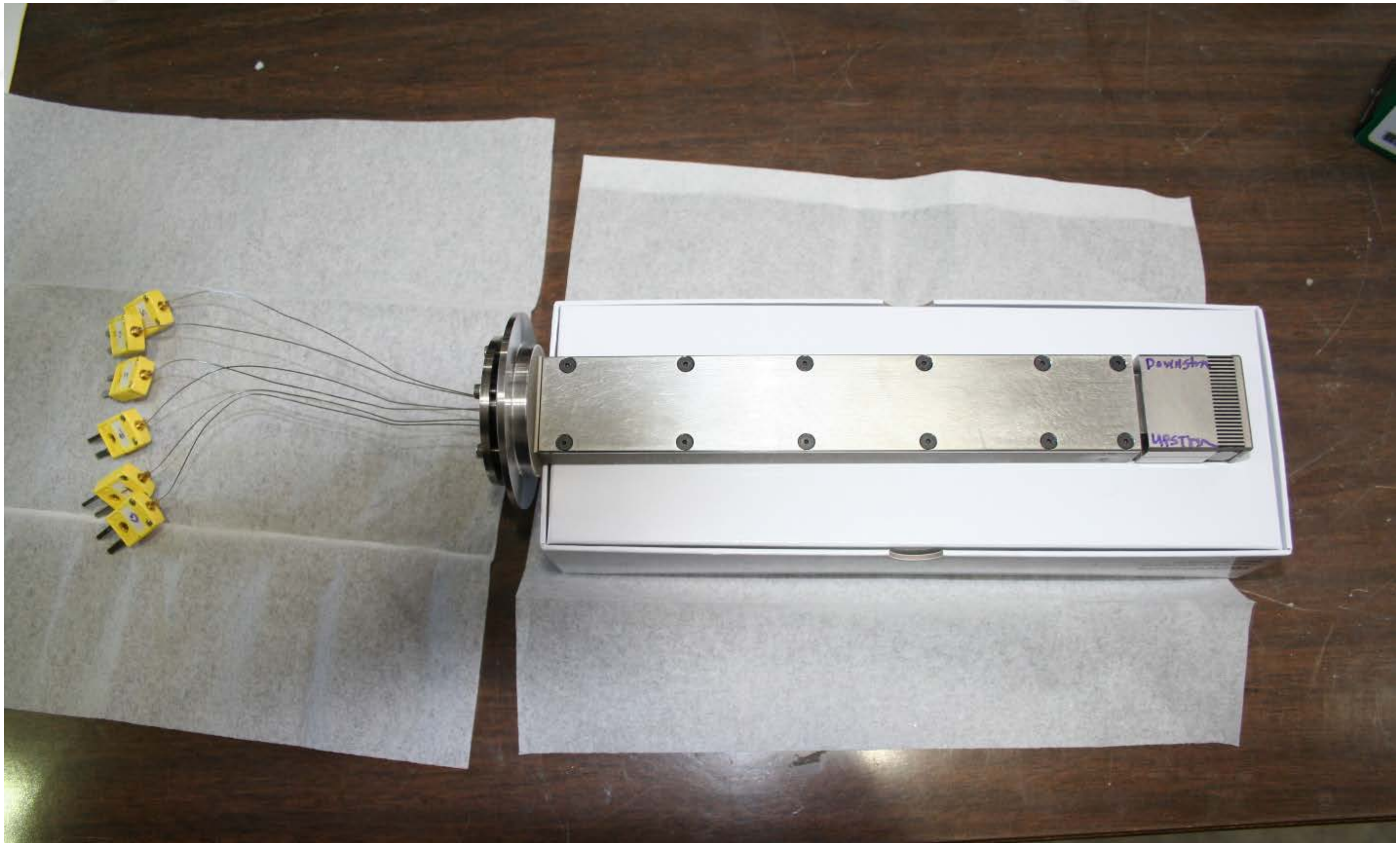
- 6.5 day production test
- Thermal test using pressed powder Mo disks

Currently Fabricating a target for 29 mm diameter disks

12 mm Diameter Target Tests Completed in the Last Year

- 6.5 day production test
  - 6 enriched ^{100}Mo disks, 19 natural Mo disks
  - ~8 kW average beam power
  - ~ 20 Ci of ^{99}Mo, ~ 17 Ci produced in the 6 enriched disks.
- Thermal test using pressed powder Mo disks
  - Tested 4 pressed and sintered natural Mo disks from ORNL under high heat load and thermal shock conditions to verify the structural integrity of the disks.

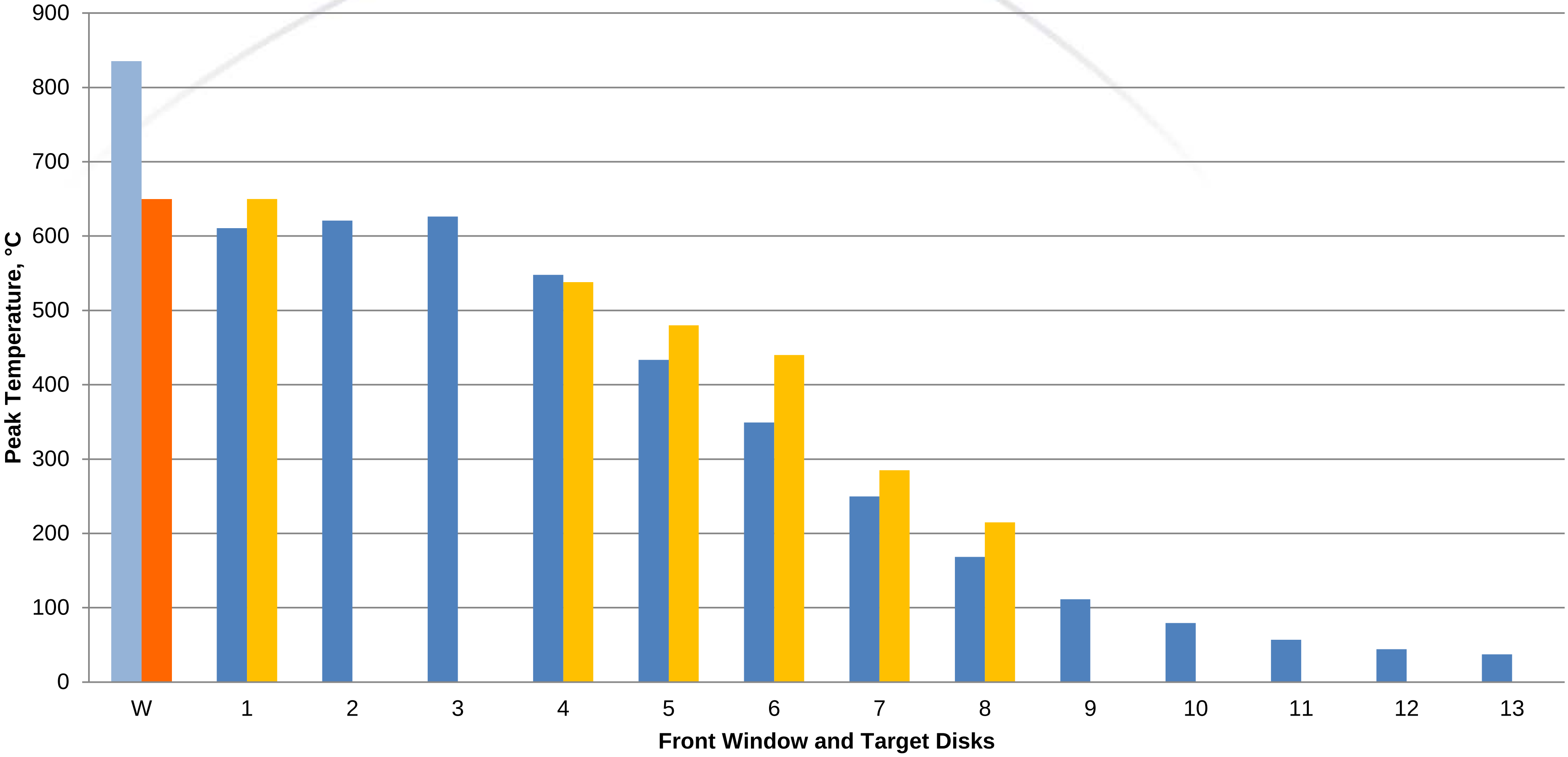
Target Assembly for the Thermal Test with Pressed Powder Natural Mo Disks



Target mounted to insertion stalk, with thermocouples

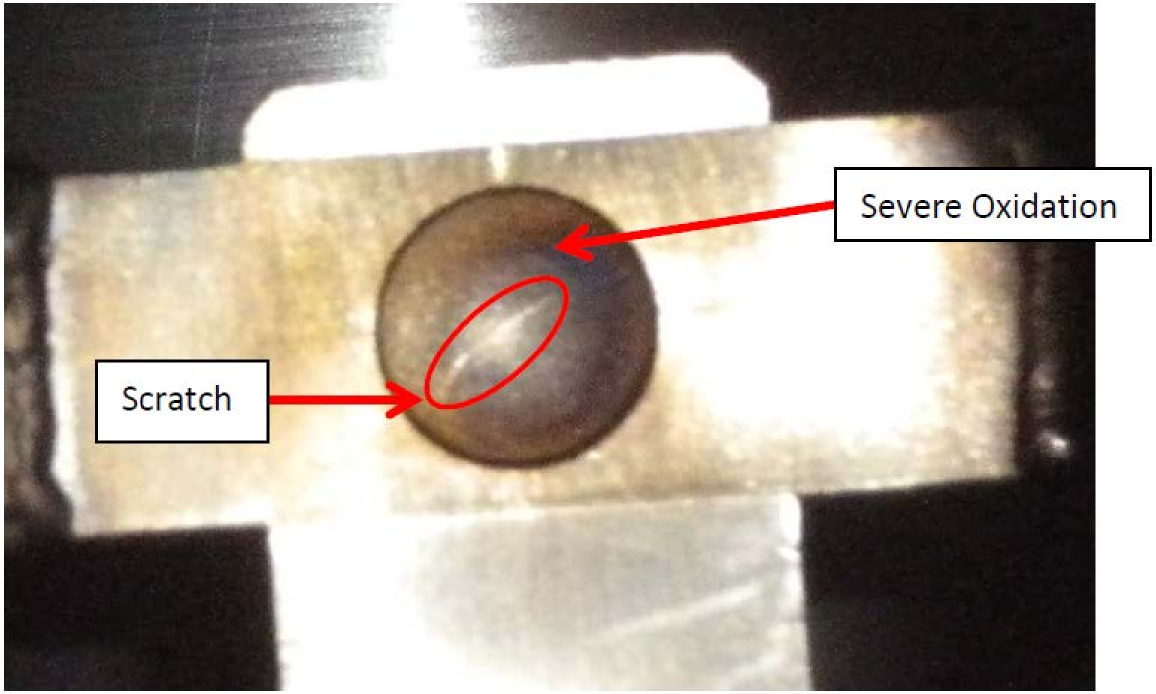
Thermal Test Results

Peak Window and Target Disk Temperature: 23MeV @ 275 psi Inlet ($\dot{m}$ = 94 g/s)



Calculated temperatures in blue, along with measured temperatures in orange. For the window, the light blue bar is the calculated value, the dark orange bar the IR camera measurement.

Front Window Oxidation

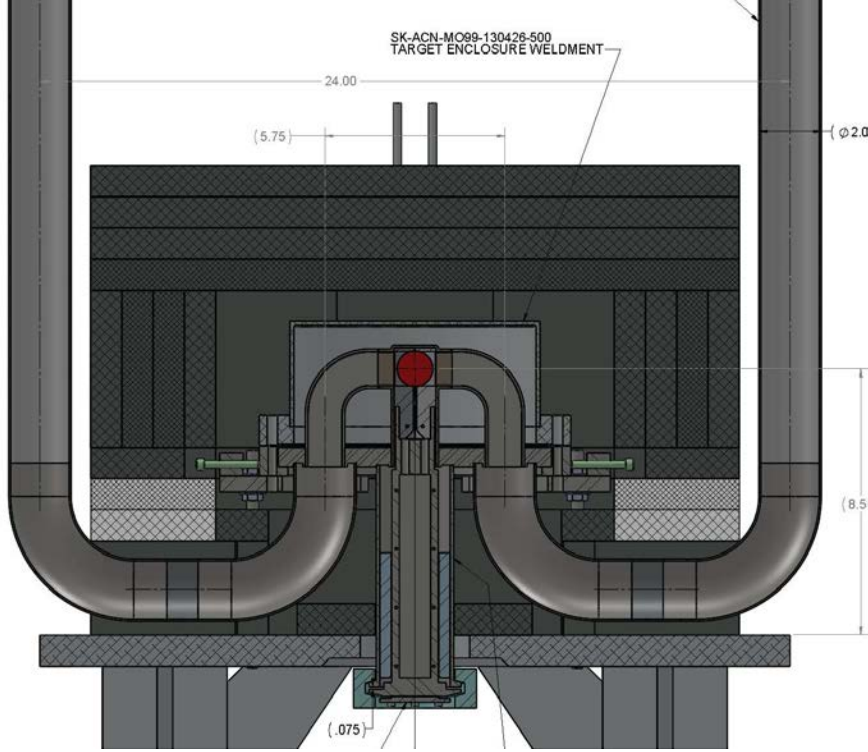


- Oxidation of the beam window has been complicating the IR temperature measurements.
- A scratch on the window caused during installation is also complicating our beam profile measurements.
- These can be mitigated with proper handing and leaving the window under vacuum after irradiation.

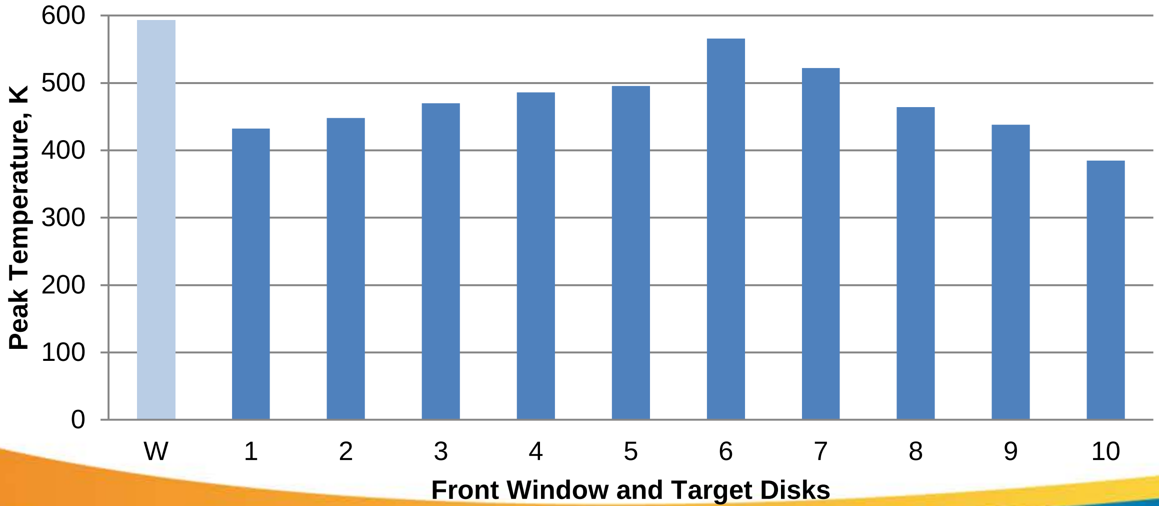
29 mm Target System for Testing at ANL

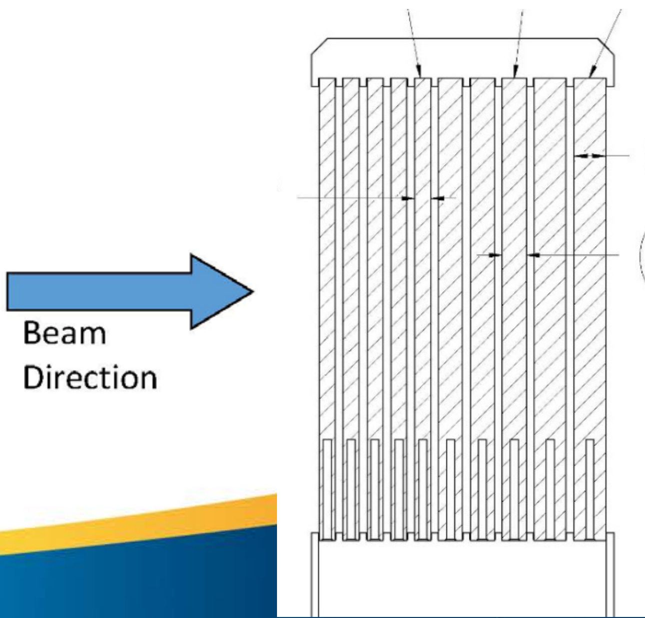
A larger diameter target will be needed for production at the beam powers being considered.

The new target design also varies the target thickness, which creates a more even power profile through the length of the target.



Peak Window and Target Disk Temperature: 35 MeV @ 285 psi Inlet ($\dot{m}$ = 0.116 kg/s)





Testing a Production Prototype Blower System

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A production prototype helium blower system is undergoing testing at LANL.

We have completed two 7-week "production" runs.

The production prototype blower system has ~ 3 times the mass flow rate for cooling targets as the system we installed at ANL.

Summary

- LANL is partnering closely with NNSA and the other National Laboratories to help develop the commercial domestic production of ^{99}Mo without the use of HEU.
- Under the M^3 ^{99}Mo Program, we are currently supporting NorthStar Medical Radioisotopes and SHINE Medical Technologies.
- Leveraging the unique capabilities of the National Laboratories to facilitate the domestic production of ^{99}Mo.

Additional Slides

Future Thermal-Hydraulics Modeling

- SUPO validation work to continue
  - Heat transfer coefficient correlation
- Power vs. bubble size relationship determined from SUPO analysis will be used in a revised solution vessel model.
  - Coupled MCNP-Fluent calculations
  - Solution vessel parameter study
    - Operating power
    - Vessel aspect ratio
    - Cooling tube number
    - Inlet coolant temperature
  - Transient coupled calculations
    - Gas-liquid interface tracking
  - Heat transfer coefficient correlation for use in a simplified system model
- Argonne Bubble Experiment model update