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Title: Los Alamos National Laboratory Support for Commercial U.S. Production of 99Mo without the Use of Highly Enriched Uranium

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Los Alamos National Laboratory Support for Commercial U.S. Production of ^{99}Mo without the Use of Highly Enriched Uranium

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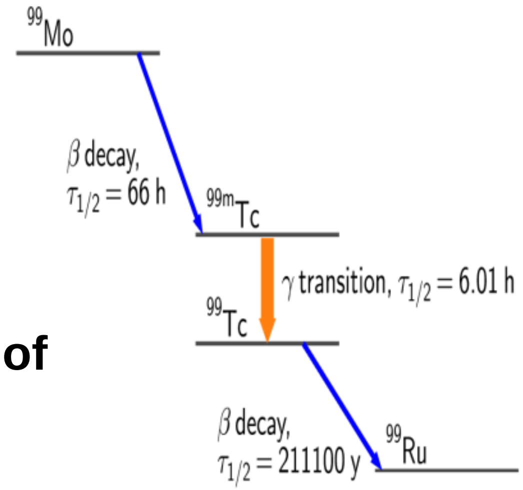
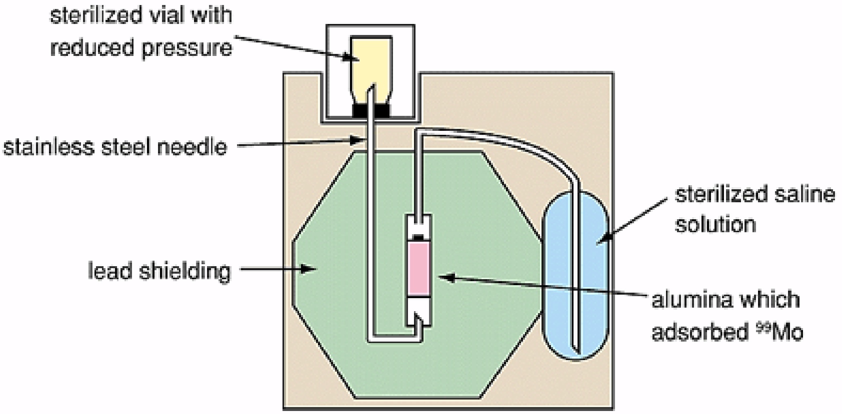
Use and Distribution of ^{99m}Tc

- **^{99m}Tc is used for diagnostic imaging.**
  - Decays with a 141 keV internal transition (IT) γ.
  - 6-hour half life.
- **^{99m}Tc is the daughter product of the longer lived ^{99}Mo.**
  - 66-hour half life.
  - Allows distribution using radionuclide generators.
- **The source of over 99% of the world's supply of ^{99m}Tc from ^{99}Mo is produced from the fissioning of ^{235}U.**
- **Most of this production uses Highly Enriched Uranium (HEU) targets.**



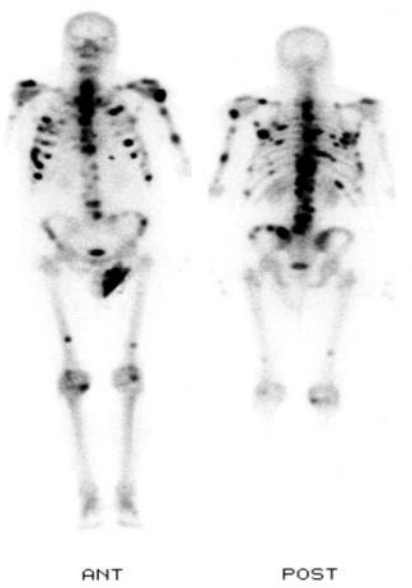
Integrated SPECT/CT Imager



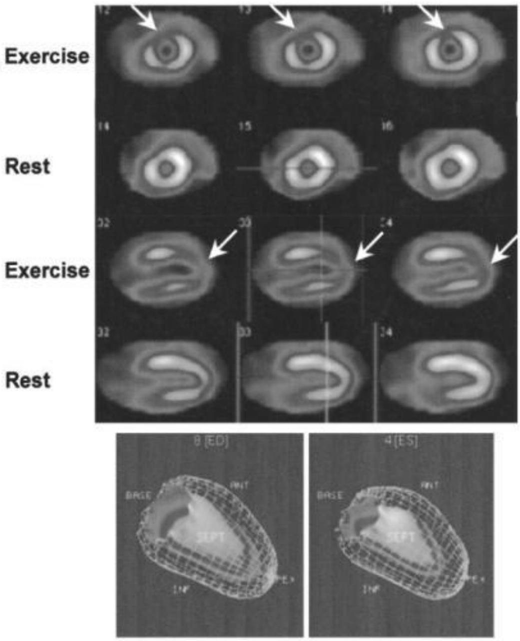
an example of the structure of a generator on the market

Diagnostic Imaging with ^{99m}Tc

- ~80% of diagnostic imaging procedures in nuclear medicine use ^{99m}Tc, ~50,000 procedures every day in the US.
- The isotope's short half-life and excellent binding properties make it uniquely suited for medical imaging procedures.
- Also due to this short half-life, it must be continuously produced to meet the medical community's requirements.
- Recent production disruptions have seriously impacted the supply of this critical medical radioisotope.



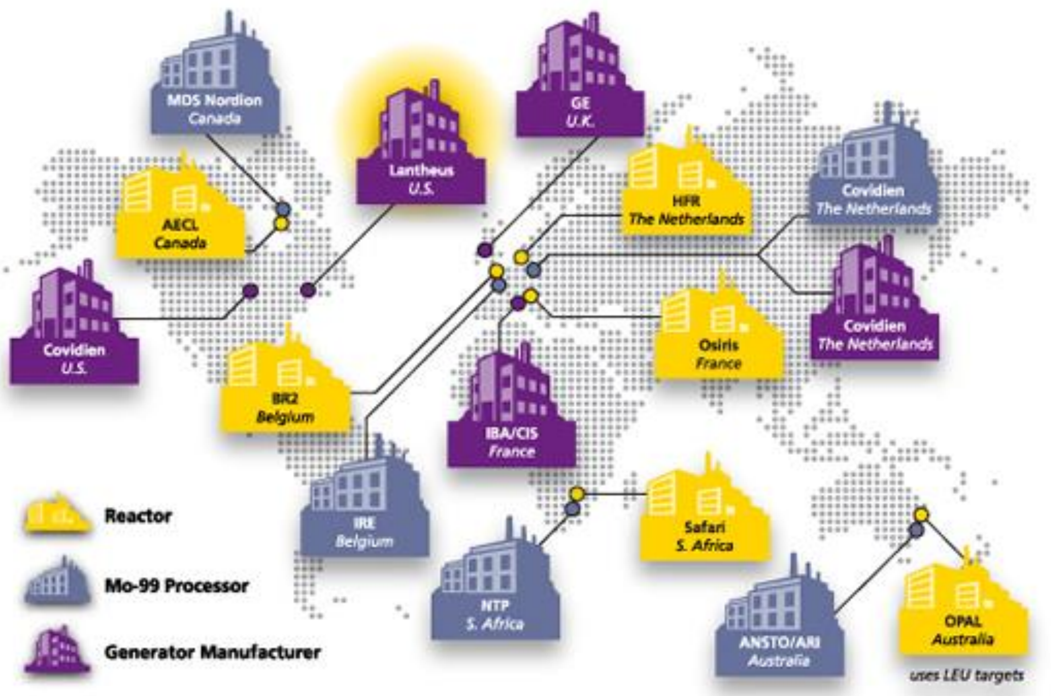
Bone Imaging~ 16%



Heart Imaging ~60%

Global Production of ^{99}Mo

Current Global State of ^{99}Mo Production
http://www.radiopharm.com/SupplyUpdate/index.html



Past major suppliers of ^{99}Mo to the US:

NRU, Chalk River (60%)

HFR, Petten (40%)

NRU and HFR produce 95% of the world supply of ^{99}Mo.

- **^{99}Mo is currently produced in nuclear reactors as a fission product from HEU targets (fission yield is 6%).**
- **There is currently a critical shortage of ^{99}Mo in the US.**
  - The US is completely reliant on foreign suppliers for ^{99}Mo.
  - Two major production reactors are currently offline.
  - The crisis is likely to continue or worsen due to the age of the major production reactors and the lack of alternatives.

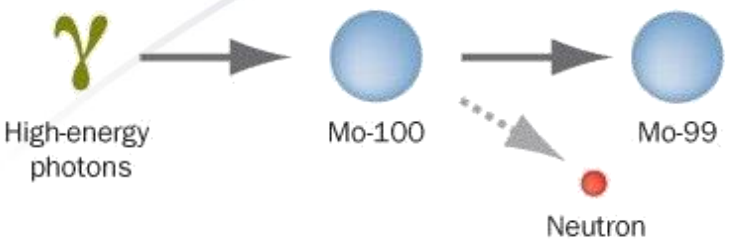
M^3 Effort in ^{99}Mo Production

- **NNSA Office of Materials Management and Minimization (M^3) effort for production of ^{99}Mo.**
  - Develop a reliable, domestic, commercial supply of ^{99}Mo that avoids a single point-of-failure and does not require the use of HEU.
  - The M^3 focus is on commercial production. Federal facilities should not be used for production or distribution. The efforts of the national laboratories are for engineering design support and proof of concept demonstrations.
- **M^3 is working on implementing projects to demonstrate the viability of non-HEU based technologies for large-scale production, which include the following technology pathways:**
  - LEU targets (mostly foreign production support)
  - LEU solution reactors (B&W)
  - Neutron capture (GE/Hitachi on hiatus)
  - Accelerator production.
    - DT accelerator driven U solution (Shine/Morgridge)
    - Electron accelerator production (NorthStar)

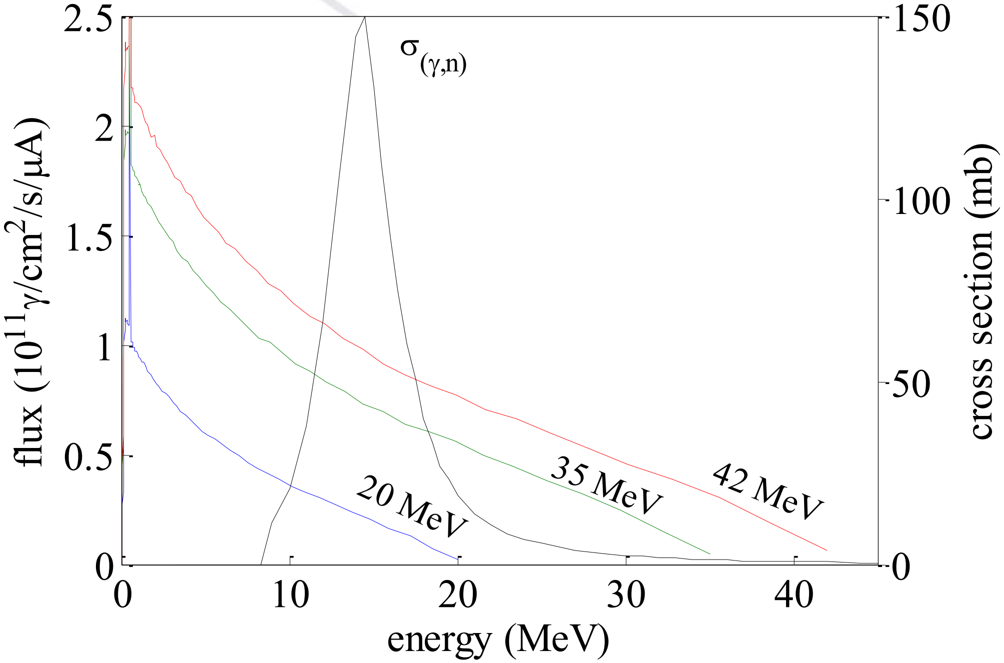
LANL Support for Domestic ^{99}Mo Production

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

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.

NorthStar Support Focus Areas

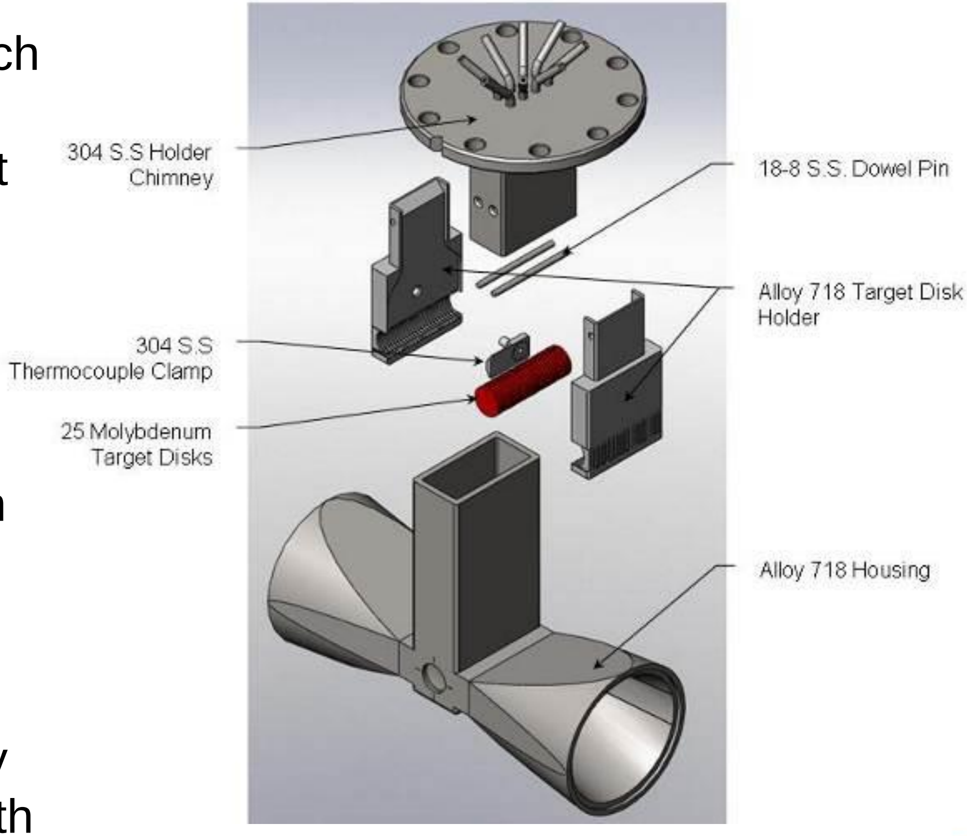
- Production and Thermal Tests at ANL
- Target Design and Testing
  - Target thermal performance
  - Production and radionuclide inventory
- Subsystem Development and Testing
  - Beam diagnostics
  - Target cooling system
  - Control systems
- Production Facility Design Support
  - Local target shielding
  - Beam line design
  - Target removal and conveyance

Scaled Accelerator Tests at ANL

Date	Test
April 2010	Water cooled target test using natural Mo targets, produced 236 µCi of ^{99}Mo.
May 2010	Water cooled target test using natural Mo targets, produced 377 µCi of ^{99}Mo.
July 2010	Water cooled production test using enriched ^{100}Mo targets, produced 10.5 mCi of ^{99}Mo.
April 2011	Once through gaseous helium cooled thermal test using natural Mo targets, 145 µCi of ^{99}Mo.
March 2012	Closed loop gaseous helium thermal test using natural Mo targets.
April 2014	Closed loop gaseous helium thermal test using natural Mo targets.
January 2015	35 and 42 MeV thermal tests at 13 kW and 7 kW, respectively. ~ 5 mm FWHM beam.
January 2015	Production Test 1: 42 MeV, 19 hours, 4.8 kW
March 2015	Production Test 2: 42 MeV, 19 hours, 7 kW
March 2015	Production Test 3: 42 MeV, 19 hours, 6 kW
May 2015	Production Test 4: 35 MeV, 24 hours, ~8 kW
September 2015	7 Day Production Test: 42 MeV, 156 hours, 8 kW

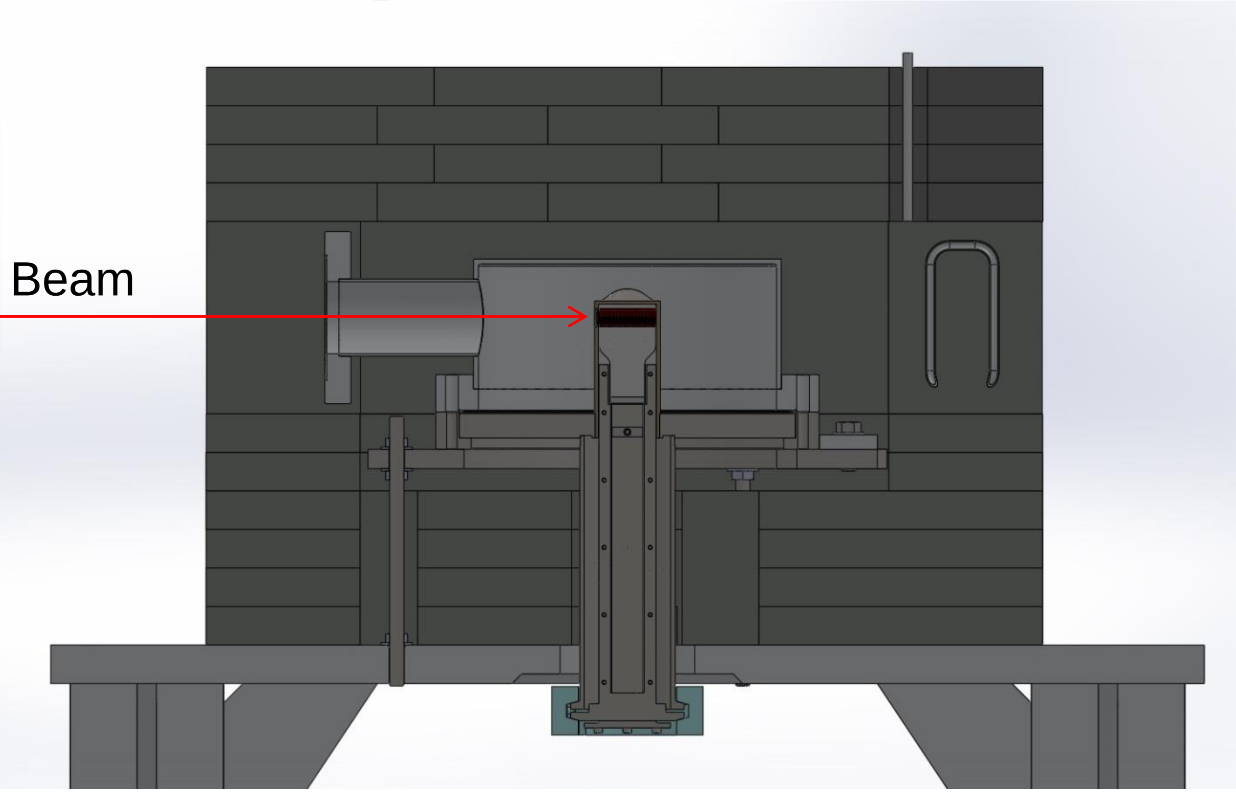
Present Target Design Concept

- Multiple Mo-100 disks held in a holder with gaps in between for coolant flow.
- The beam strikes the target from each end. Exploits target symmetry and doubles production for a given target mass.
- No x-ray converter, x-rays are produced in the Mo-100 target directly.
- The ANL test target disks are 12 mm in diameter, 1 mm thick, with 1 mm gaps, and there are 25 disks in a target.
- Production facility disks are currently 29 mm in diameter, 0.5 mm thick, with 0.25 mm gaps.



NorthStar Target Testing at ANL

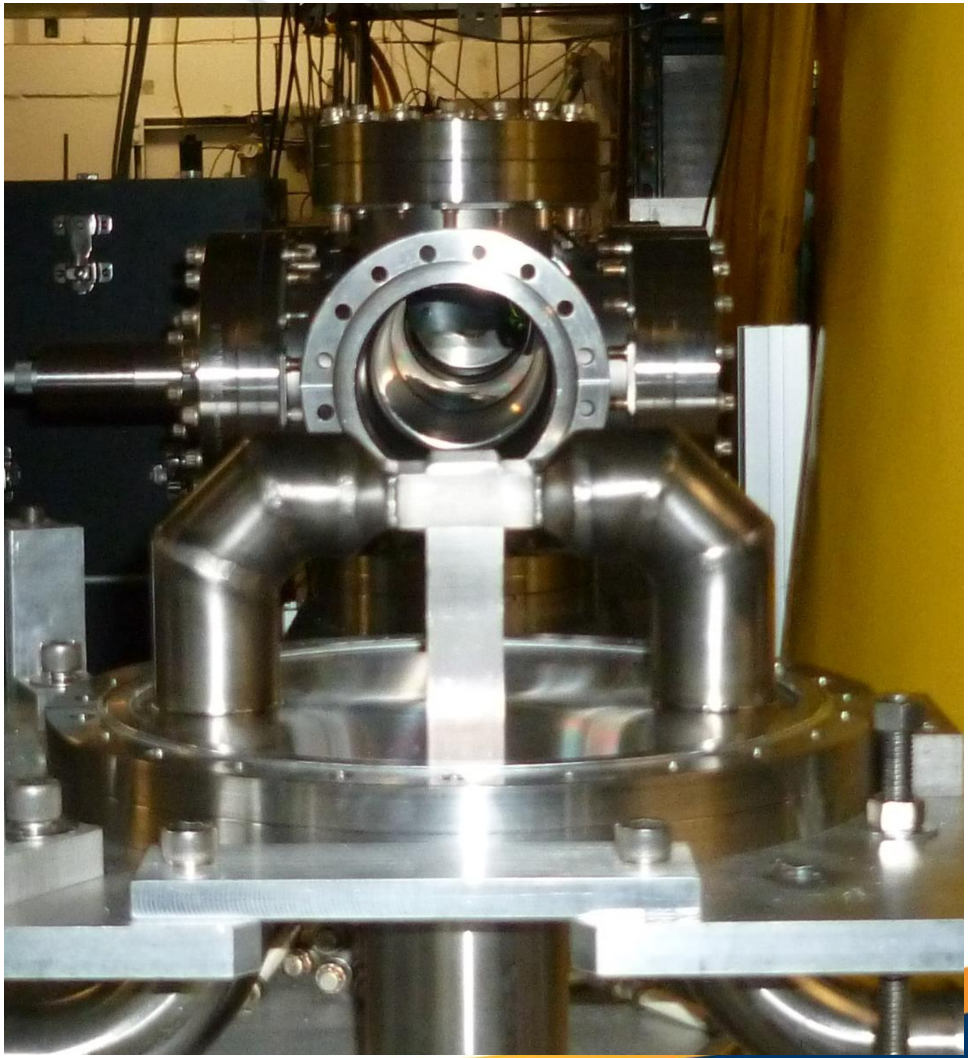
Target Side View



Single sided target for thermal and production tests at ANL

12 mm diameter, 1 mm thick disks, 25 disks in a target for testing at ANL.

April 2014 Thermal Test Setup



NorthStar Target and Helium Cooling Loop Design Installed at ANL

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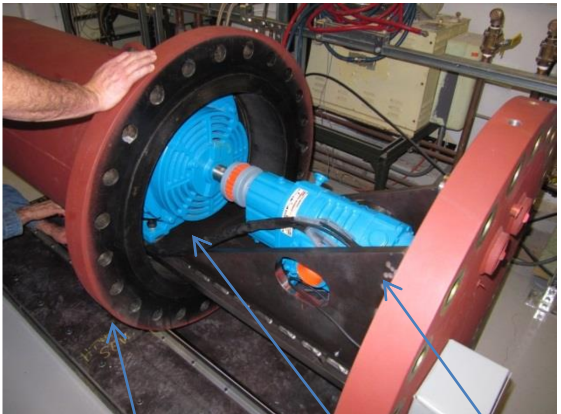
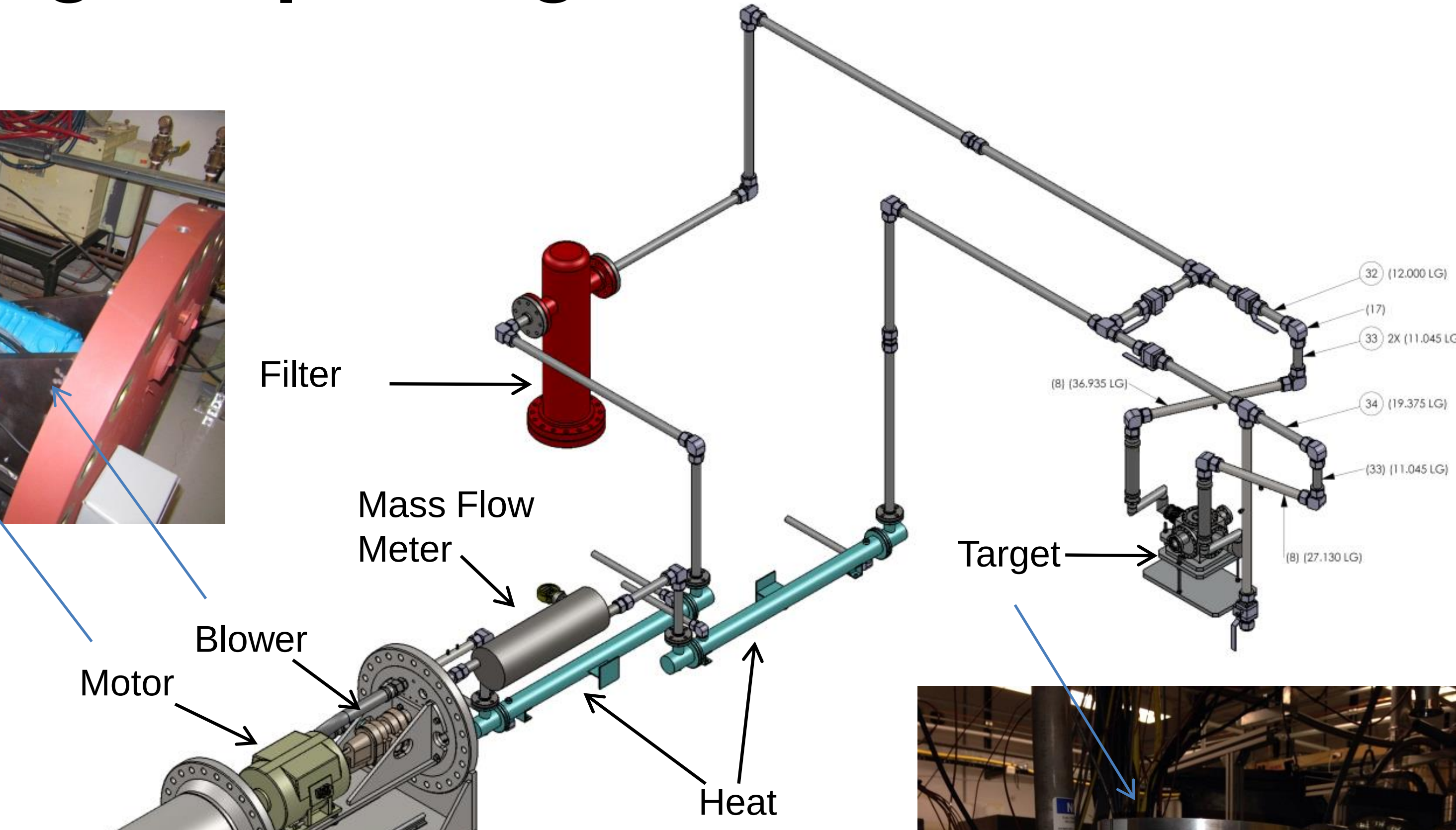
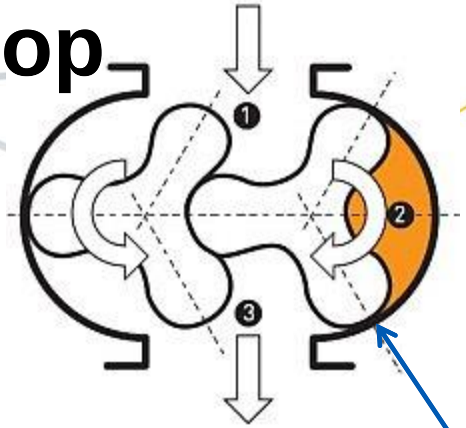
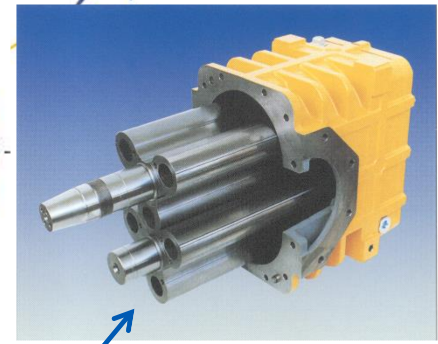




Gaseous Helium Flow Loop Using a Roots Blower

The roots blower is used to move the He through the loop and across the targets. The PV is used to increase the base pressure of the system to 300 psi.



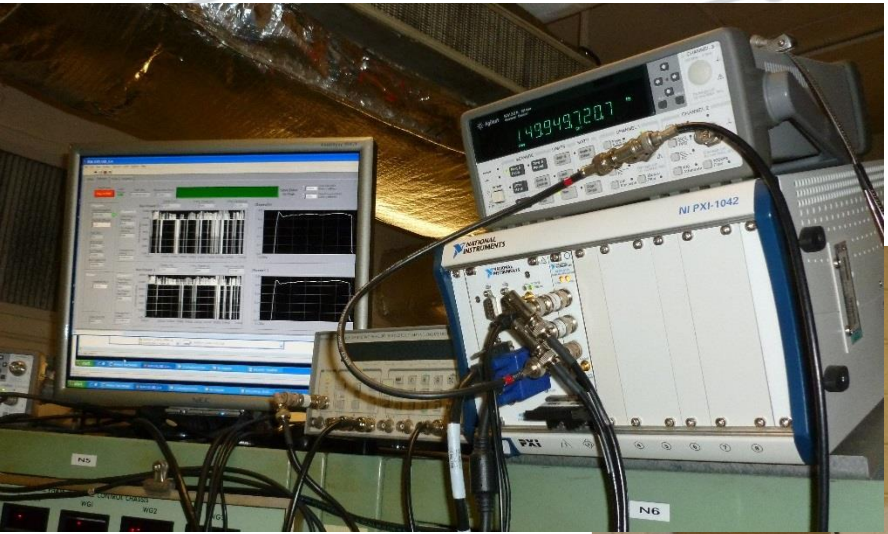


Pressure Vessel (PV)

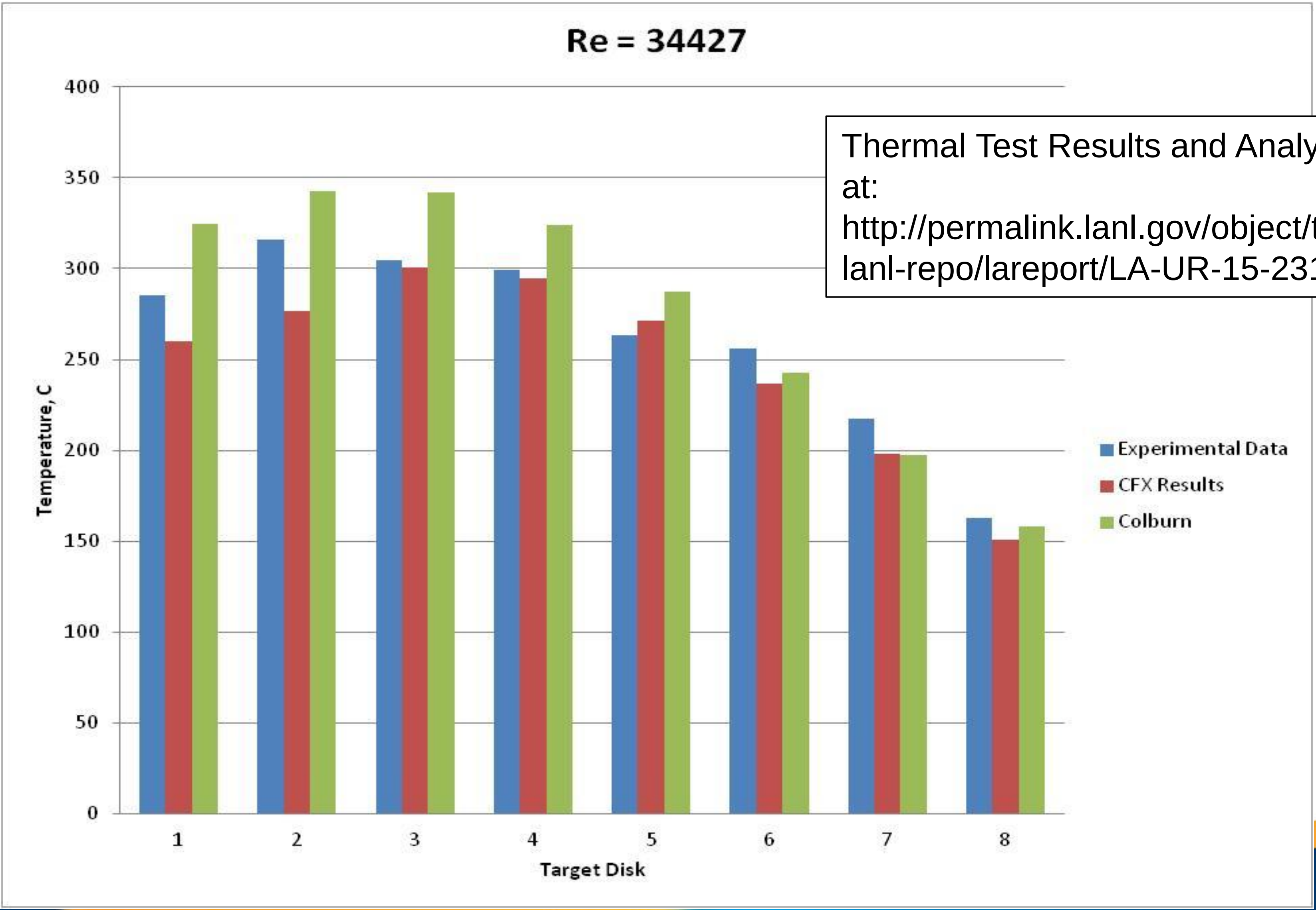
Electric motor

Roots Blower

January 2015 Thermal Test



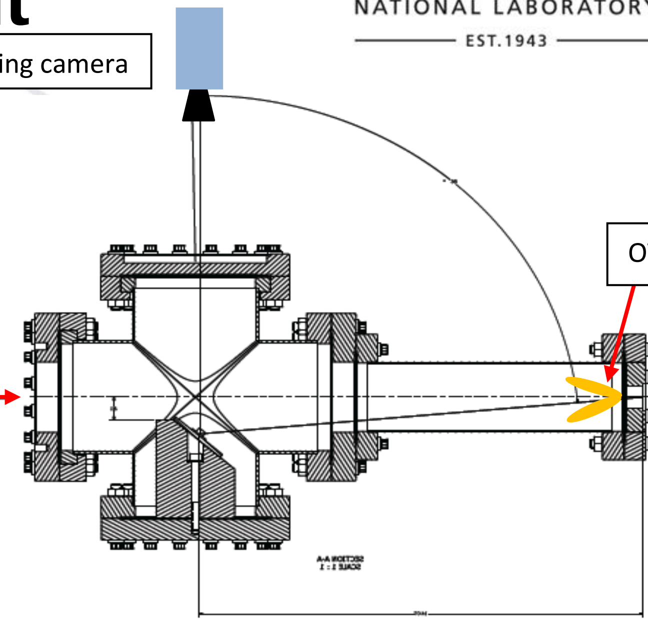
Target Thermocouple Data from the January 2015 Thermal Test 35 MeV, 13 kW beam, 290 psi inlet, 97 g/sec

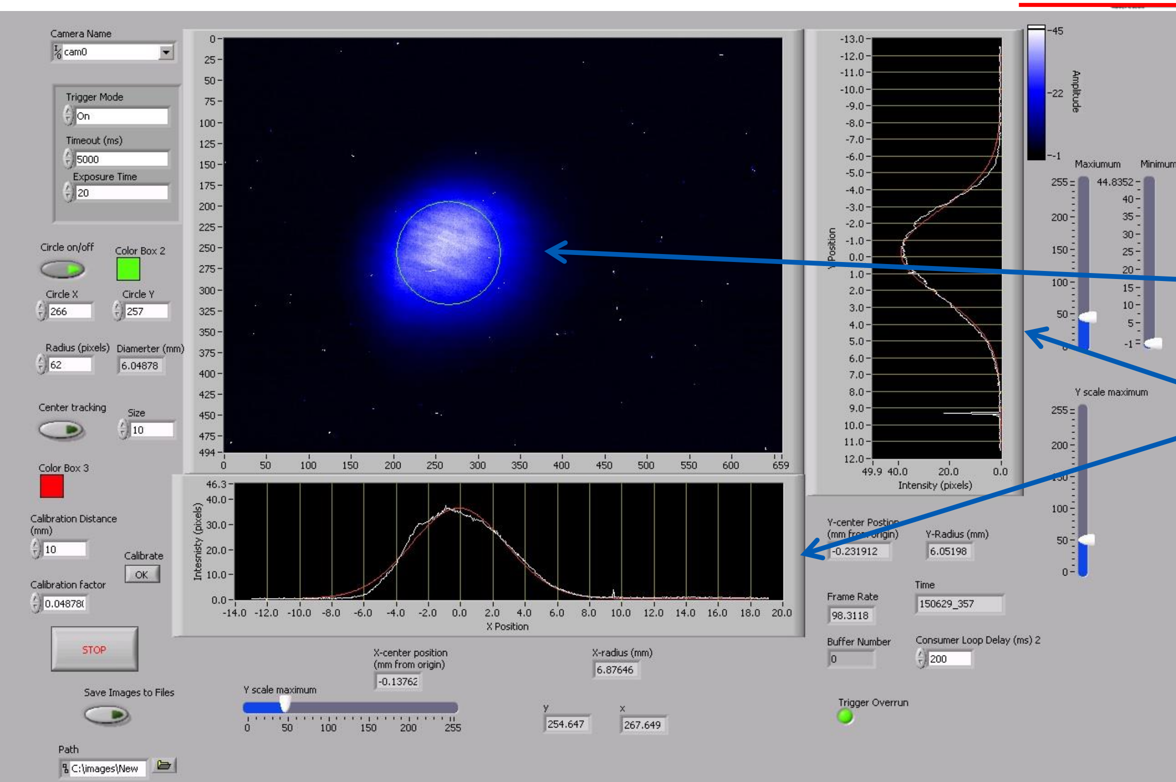


Thermal Test Results and Analysis available at: http://permalink.lanl.gov/object/tr?what=info:lanl-repo/lareport/LA-UR-15-23134

Optical Transition Radiation (OTR) Beam Position and Profile Measurement

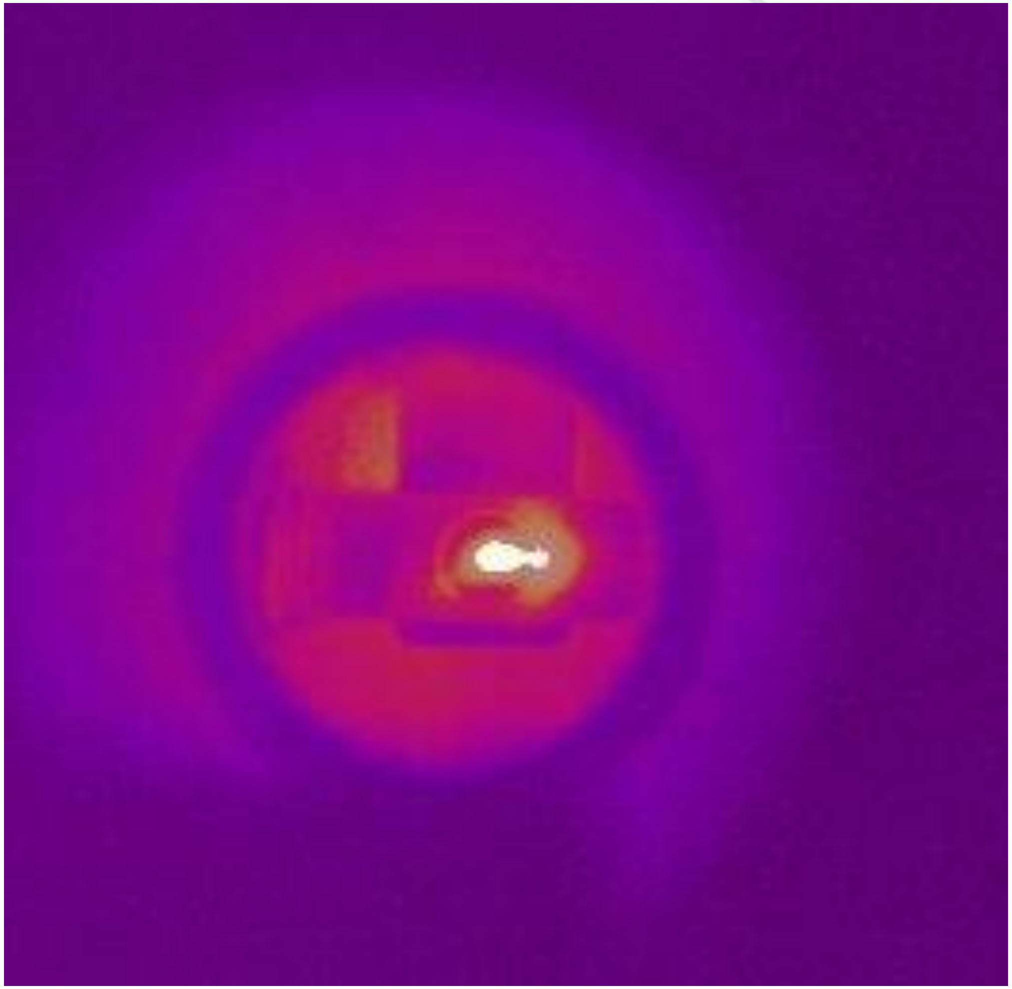
To address the beam steering and beam profile monitoring, LANL developed an OTR system looking at the exit window for measuring the ANL beam profile and position during the irradiation.





Beam image
x and y profile measurements

IR Camera Image



ENDF Photonuclear Cross Sections

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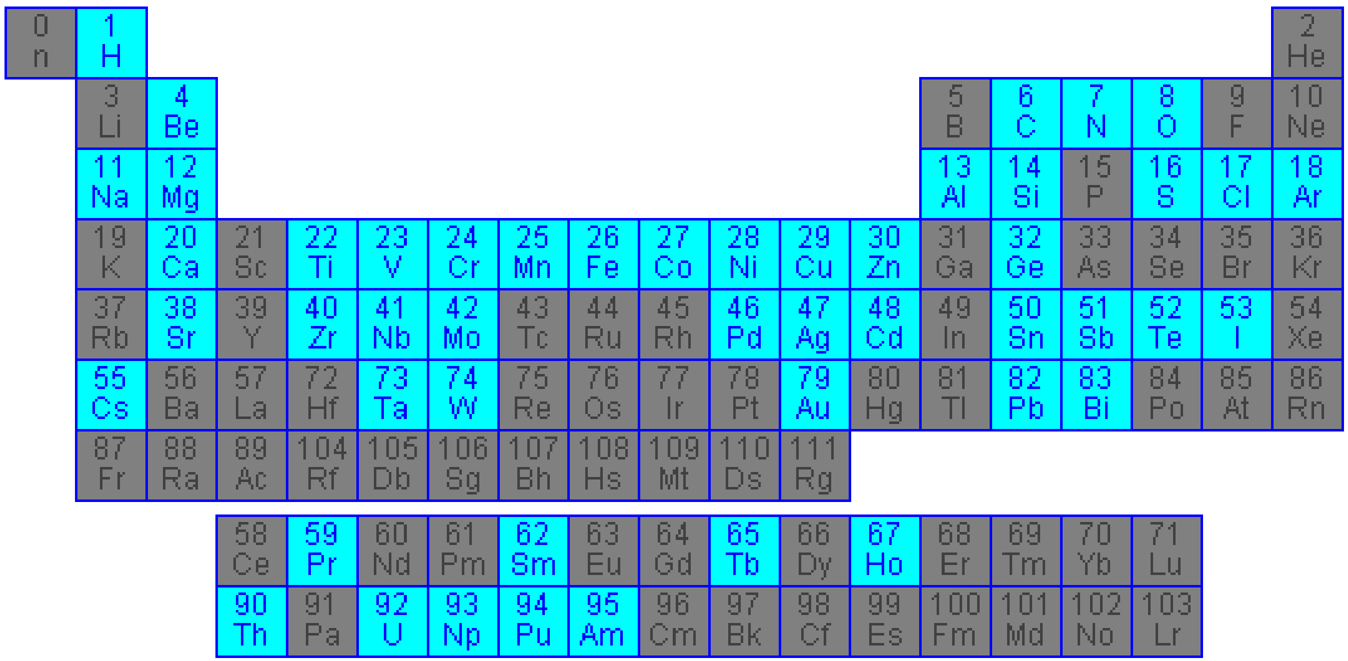
EST. 1943

ENDF/B-VII and TENDL-2012 libraries

- ENDF: 163 targets and 4335 reactions
- Elements in ENDF shown in blue in the figure below.

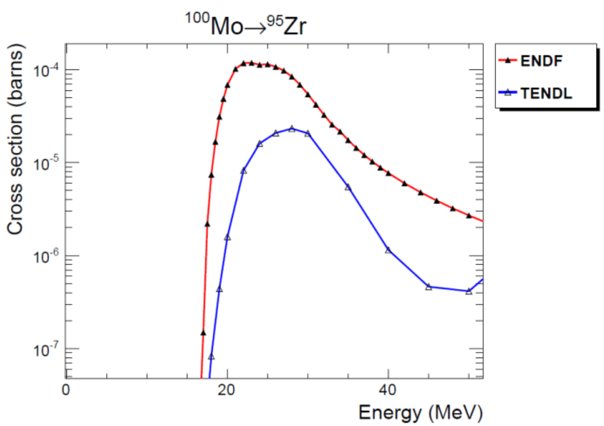
TENDL: 2400 targets and 301,565 reactions

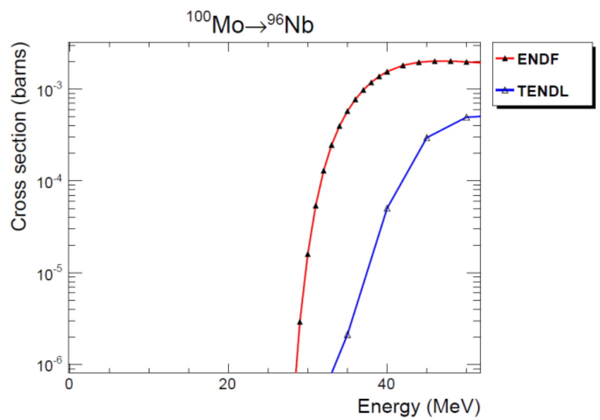


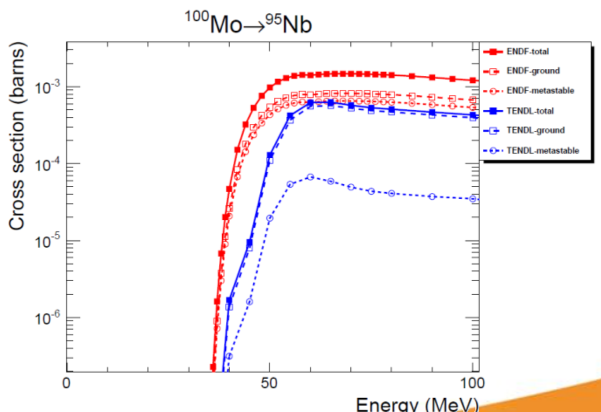
From http://www.nndc.bnl.gov/sigma/

Libraries and Side Reactions

- TENDL cross sections are generally lower than ENDF
- More side reaction and impurity activation analysis in future
- Photonuclear metastable production not in ENDF
- Half-lives: Zr-95 64 d, Nb-96 23 h, Nb-95 35 d, Nb-95m 3.6 d







Potassium Photonuclear Evaluation

From T. Kawano, "Evaluation of photo-induced reactions on ^{39,41}K," LA-UR 14-20531

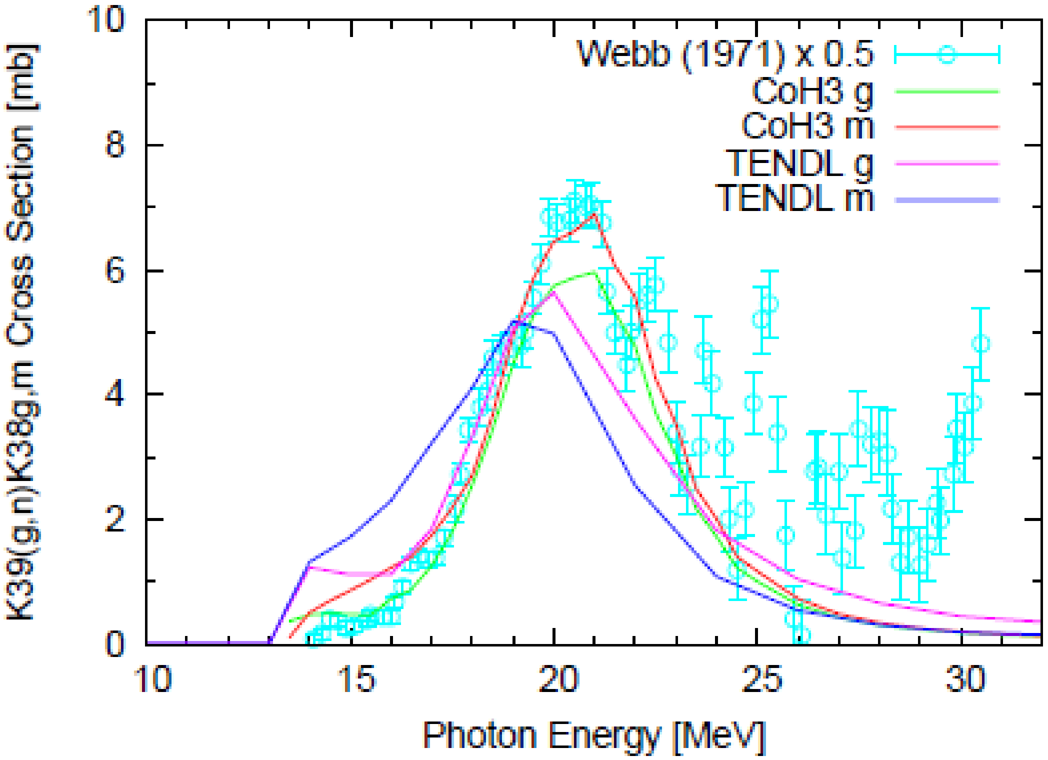
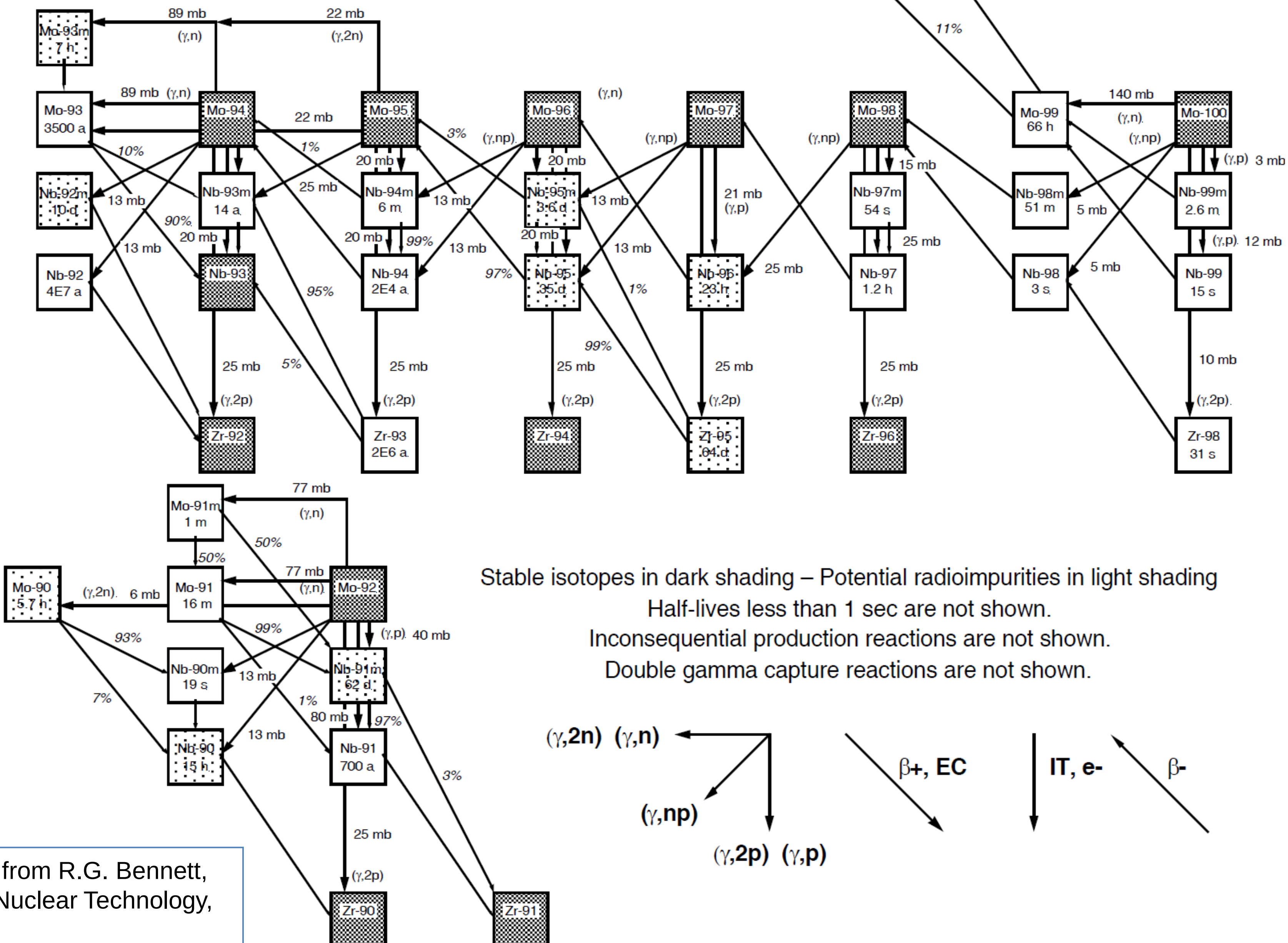


FIG. 3. Calculated ^{38m}K and ^{38g}K production cross sections for the photo-induced reaction on ^{39}K, compared with the experimental data (meta-state), as well as the evaluated data in TENDL.

Percent Abundance of Molybdenum Isotopes:

	Mo-92	Mo-94	Mo-95	Mo-96	Mo-97	Mo-98	Mo-100
Natural	14.84	9.25	15.92	16.68	9.55	24.13	9.63
Enriched Mo-100	0.60	0.23	0.40	0.81	0.36	1.69	95.9
FY10 Prod. Test	0.06	0.03	0.04	0.05	0.08	0.47	99.2



Stable isotopes in dark shading – Potential radioimpurities in light shading
Half-lives less than 1 sec are not shown.
Inconsequential production reactions are not shown.
Double gamma capture reactions are not shown.

Figure from R.G. Bennett, et al., Nuclear Technology, 1999

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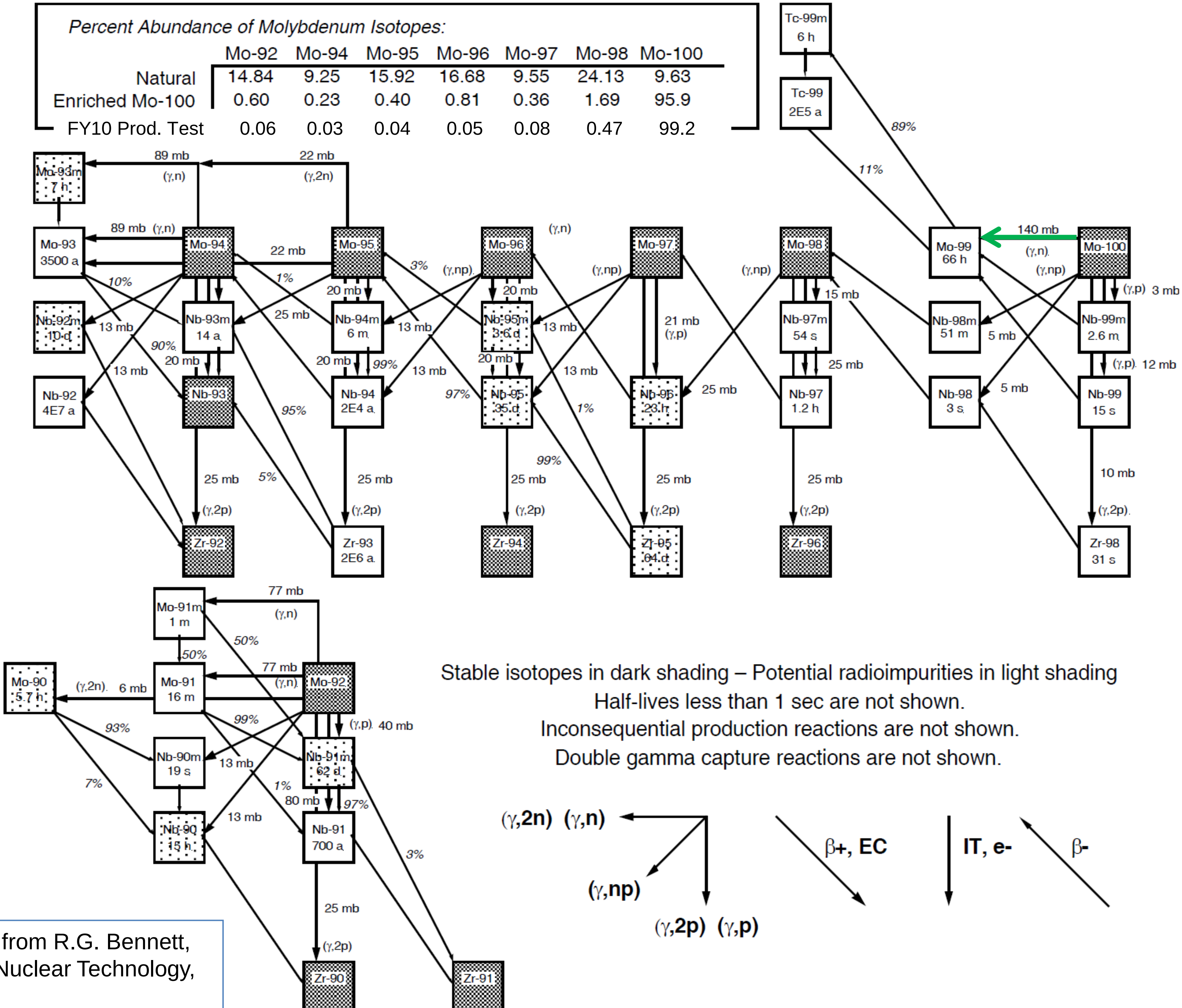

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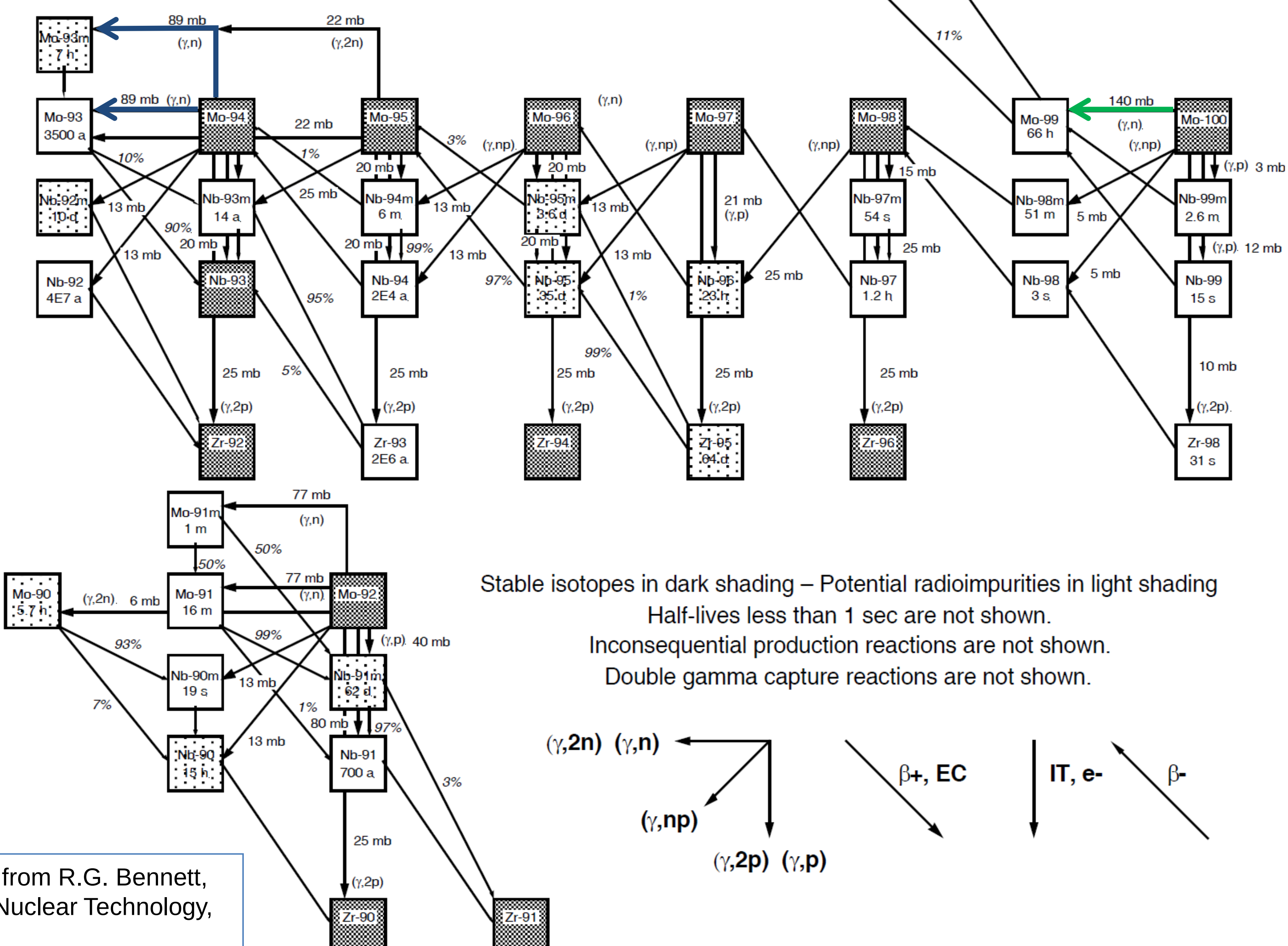

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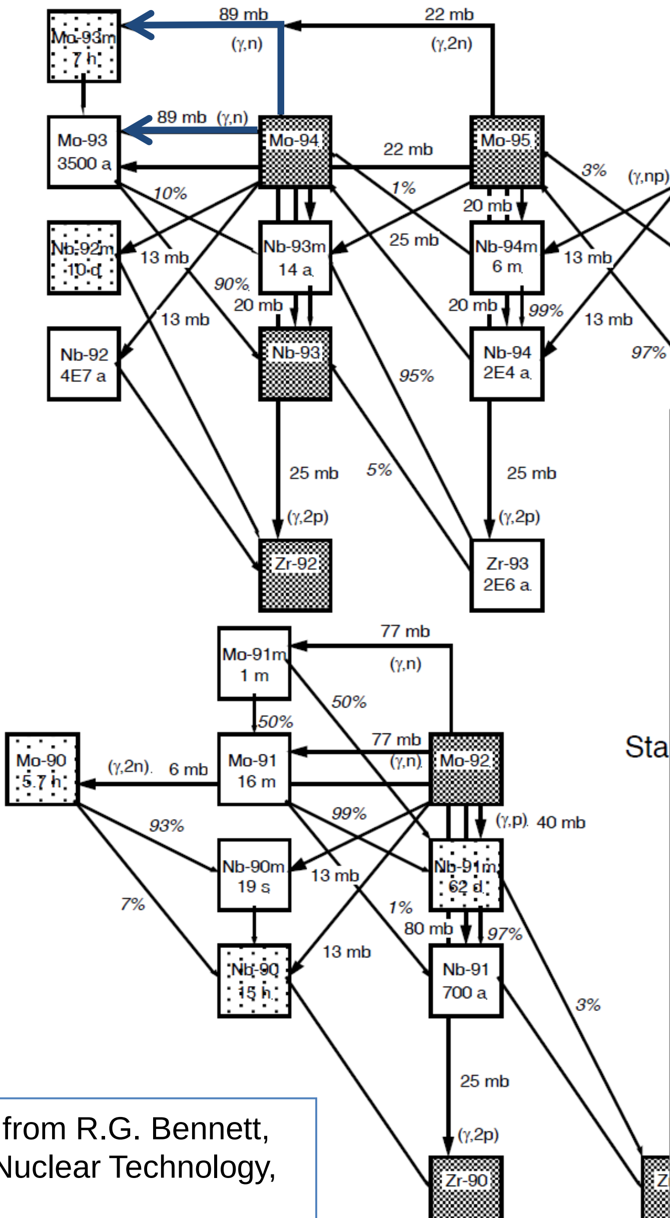

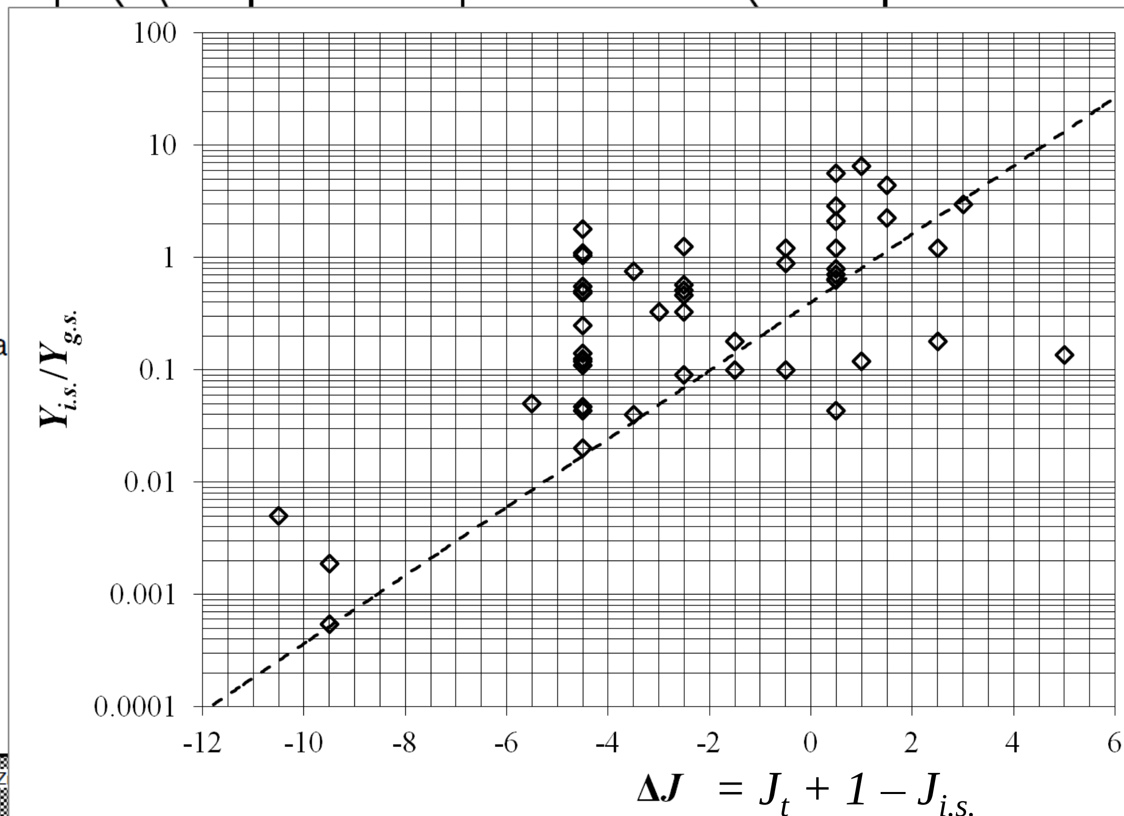

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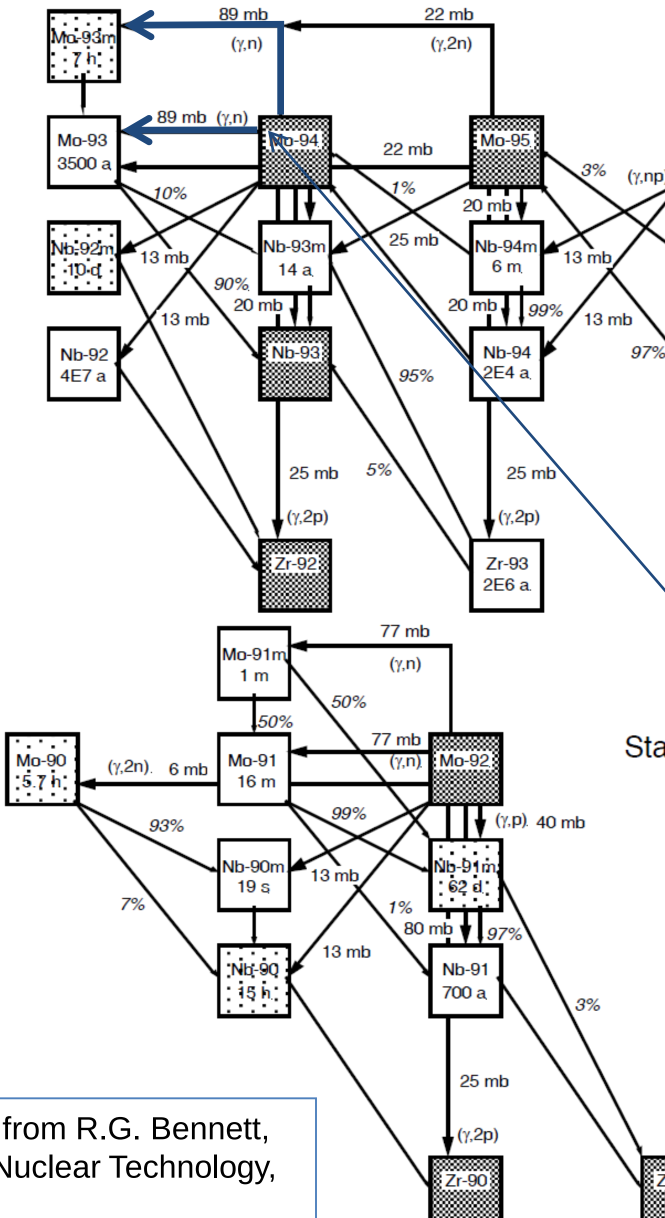

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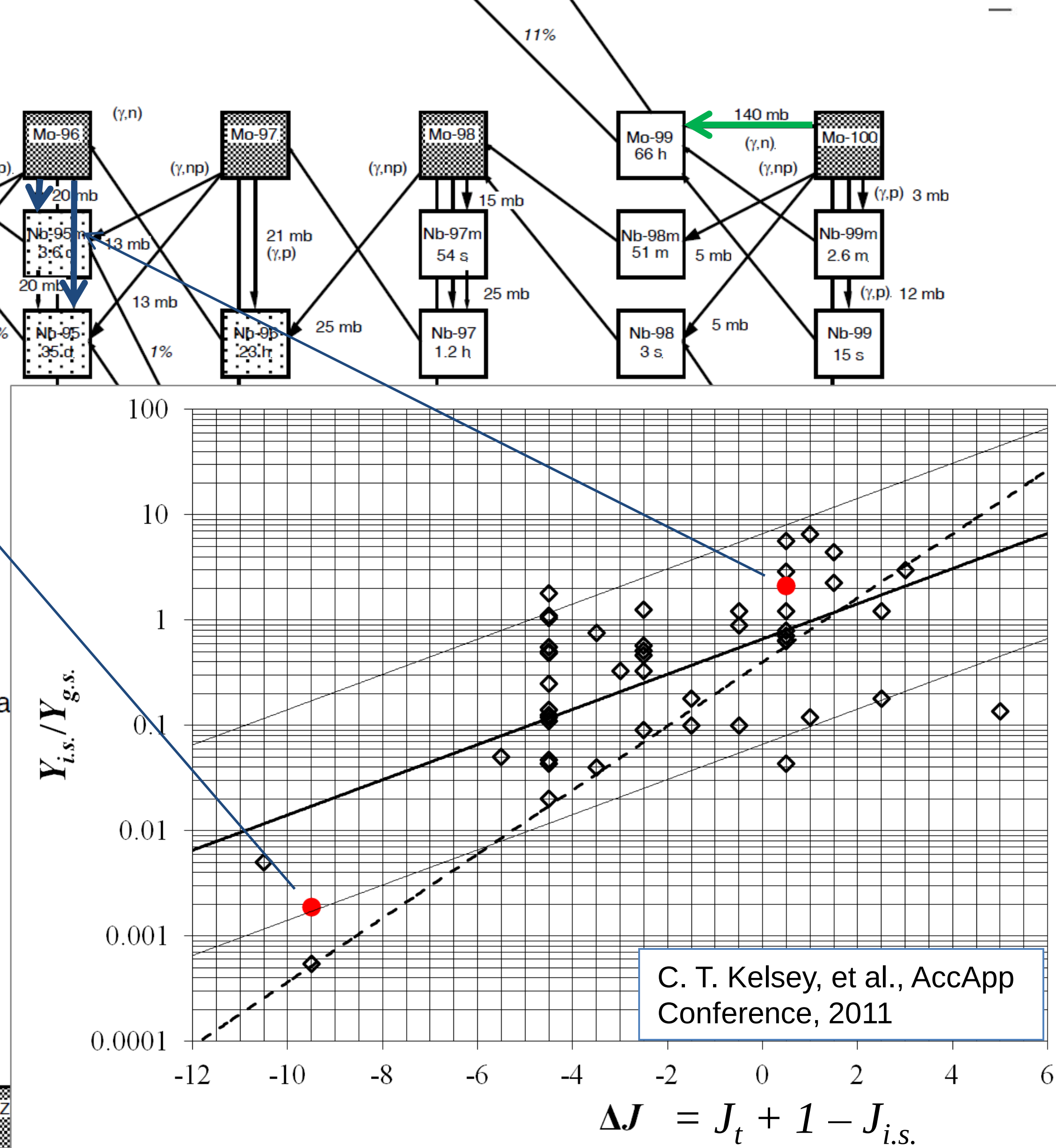
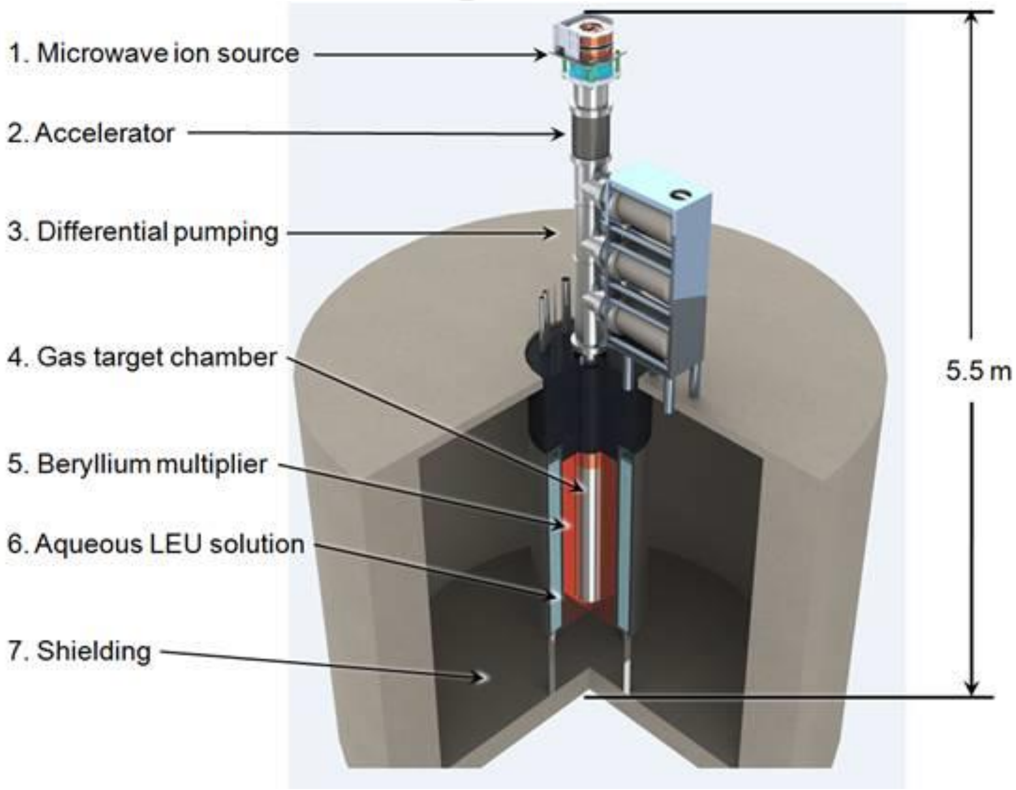


C. T. Kelsey, et al., AccApp Conference, 2011

LANL Support for SHINE Medical Technologies

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



SHINE Support Activities

- System Modeling & Design Support
  - System dynamics and reactivity modeling
  - Thermal hydraulics modeling
  - Gas nozzle design for the accelerator target.
- Irradiations and Separations Chemistry
  - Measurement and control of the total uranium concentration
- Evaluation of the Tritium Recycle Loop and Associated Systems (in partnership with SRNL)
- Zr Clad DU Target Fabrication
  - For the ANL photoneutron target for the mini-SHINE experiment.

Thermal Hydraulic Modeling

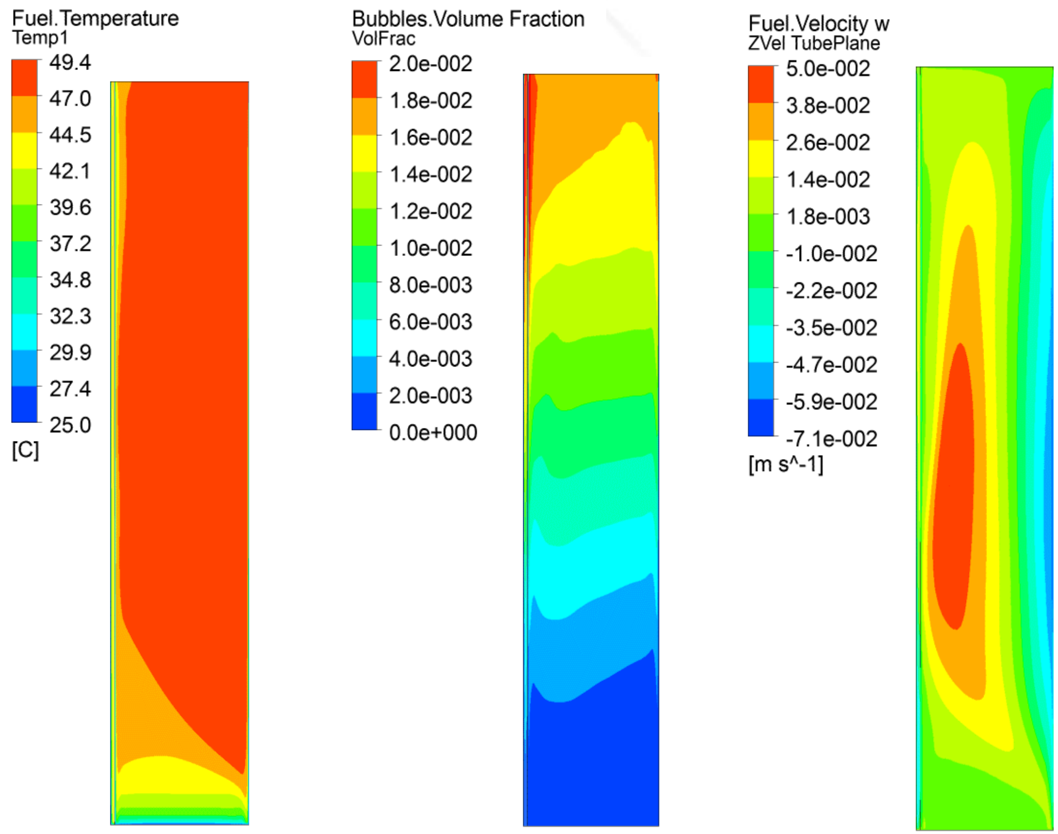
Fluid dynamics model developed to calculate steady state temperature and void fraction profiles for an externally cooled fuel solution vessel.

Computational fluid dynamics calculations performed in Ansys FLUENT.

- Heat transfer by natural convection enhanced by bubble generation
- Non-uniform volumetric heat and bubble generation profiles
- Temperature-dependent fuel and gas properties

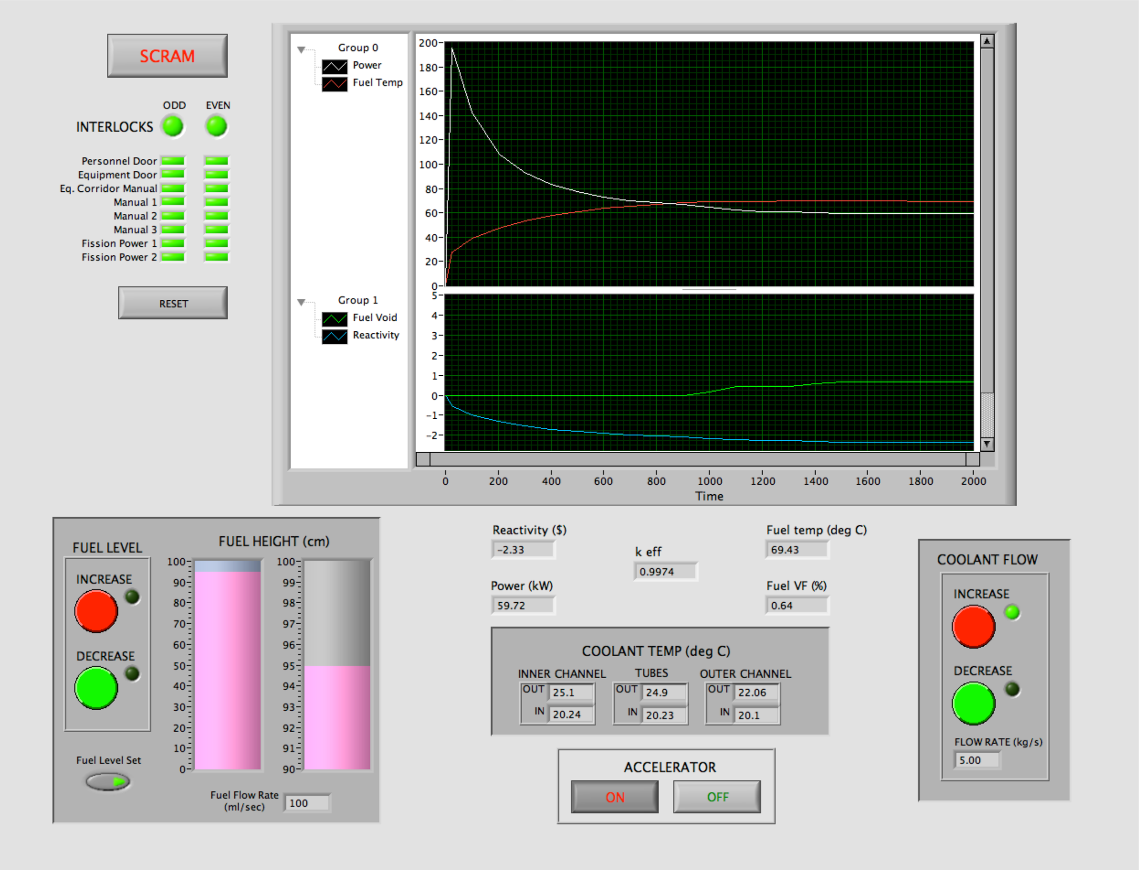
Currently iterating with reaction simulations to obtain steady-state solutions for various conditions.

- Results will be used to improve heat transfer calculations in system model.



Simulator for Accelerator-Driven System

- Implemented in LabVIEW
- Functional controls
- Strict operational sequencing and protocols enforced
- Data displays derived from DSS model
- Aids in human factors engineering of controls design
- With companion "Instructor's Screen" may be used for operator training in start-up, normal and off-normal events

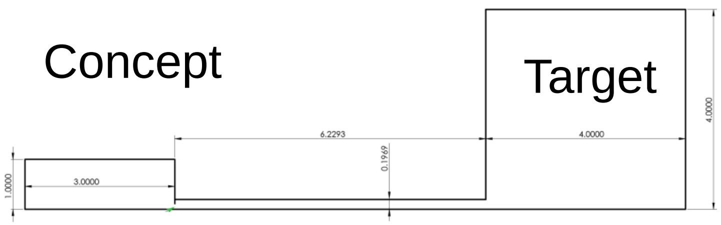


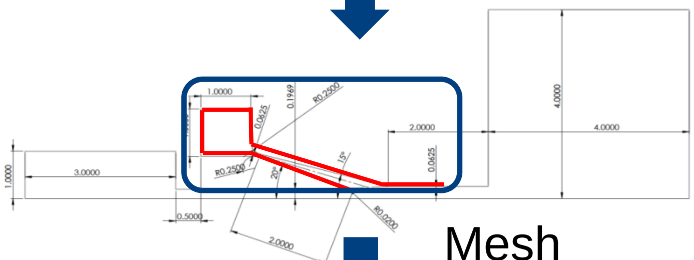
Tritium Nozzle Design

Purpose is to reduce the leakage of gas from the target to the accelerator vacuum

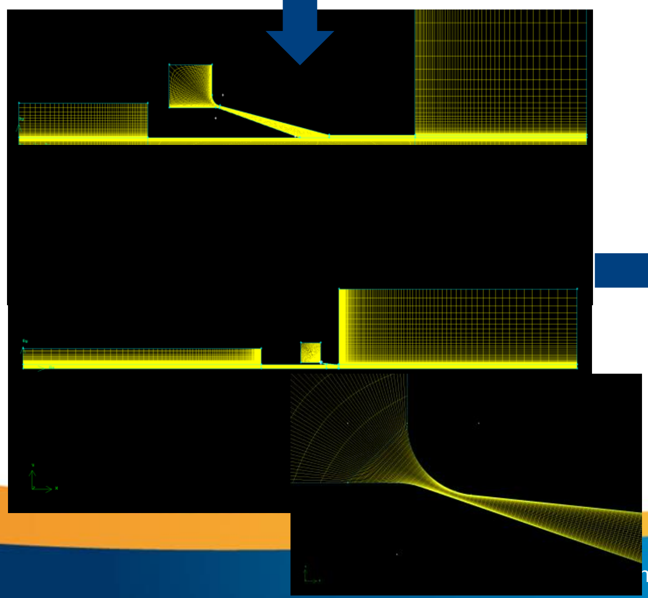
Concept

Target





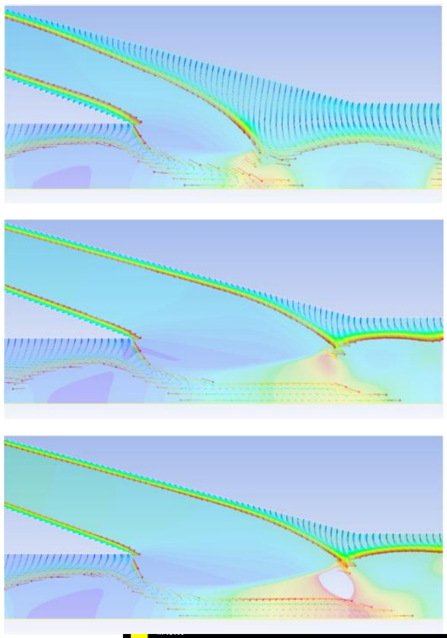
Mesh

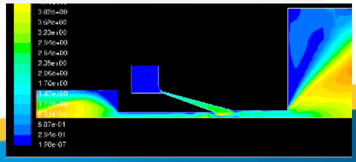


Gasses:

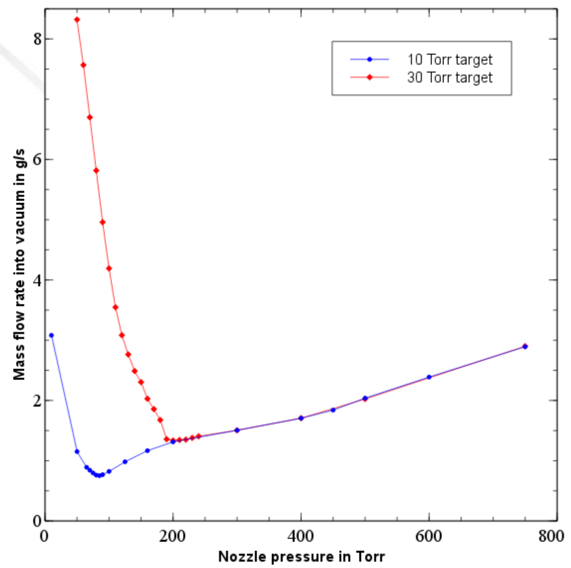
- Hydrogen
- Deuterium

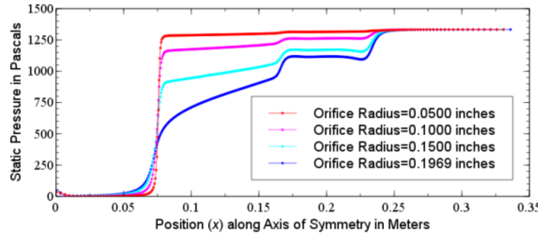
CFD





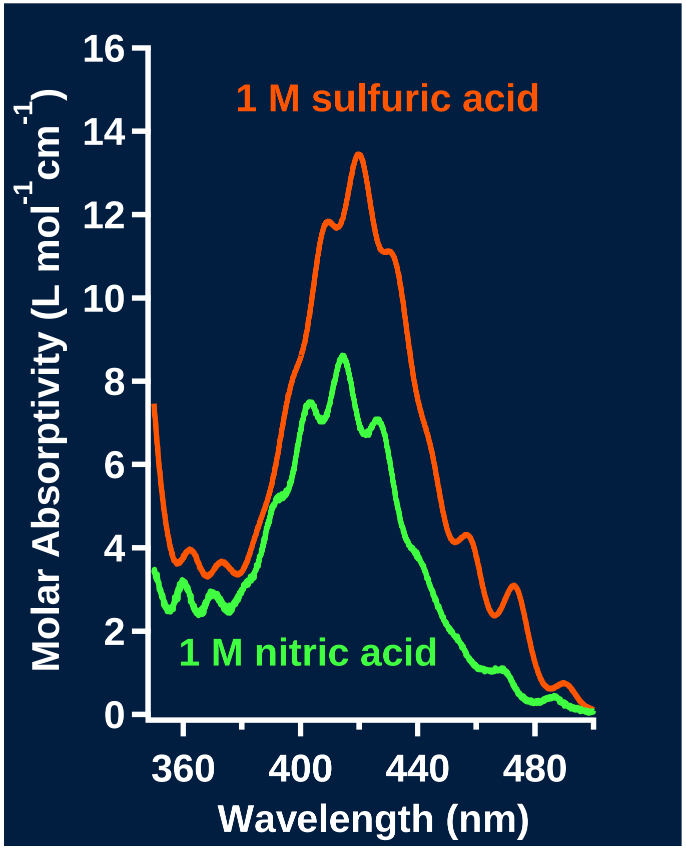
Nozzle Characterization

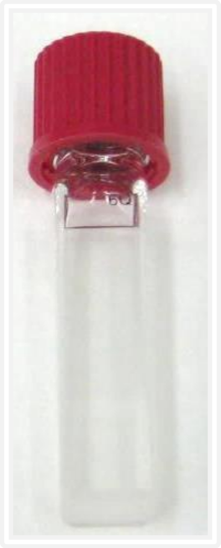


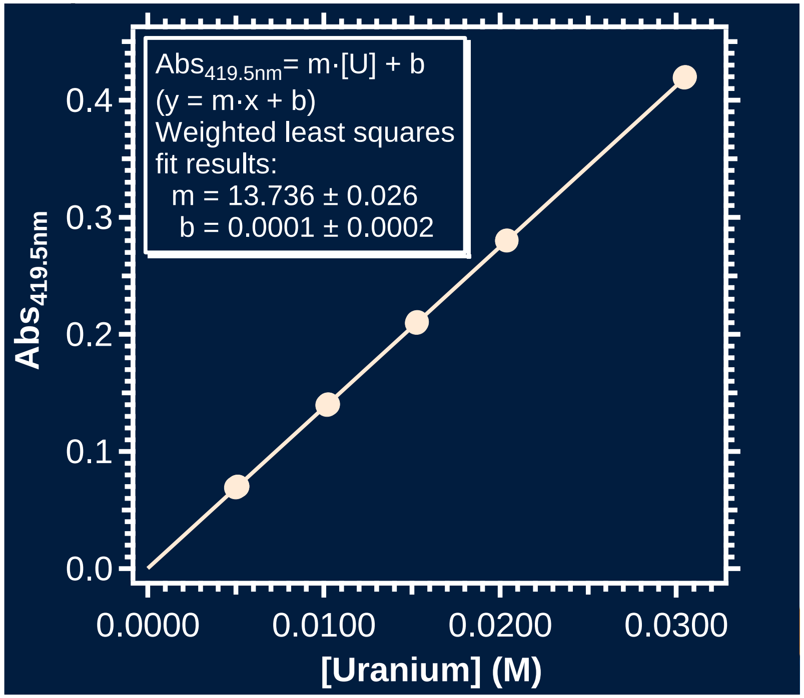


Uranium Concentration Measurement

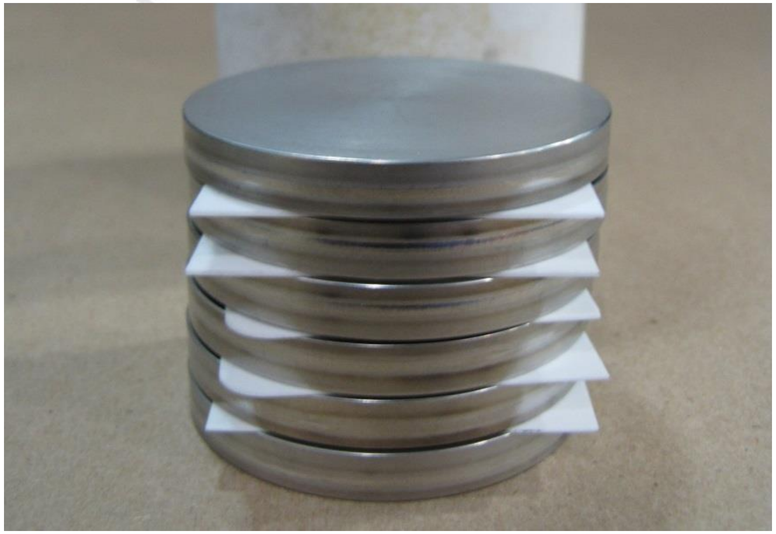
- Uranyl absorption spectroscopy – can be applied to uranium concentration measurement in solution
- A small aliquot of sample (*e.g.* 100 μL) diluted in excess of 1 M H_2SO_4 (2000 μL)
- λ_{max} (peak max, nm) and ε (molar absorptivity) vary with chemical composition







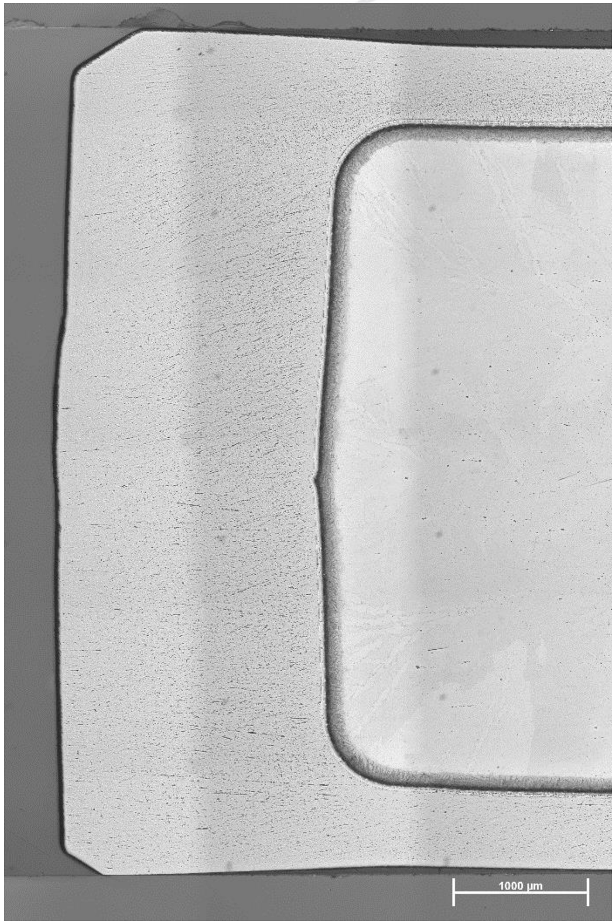
Fabrication of Zr clad DU Disks



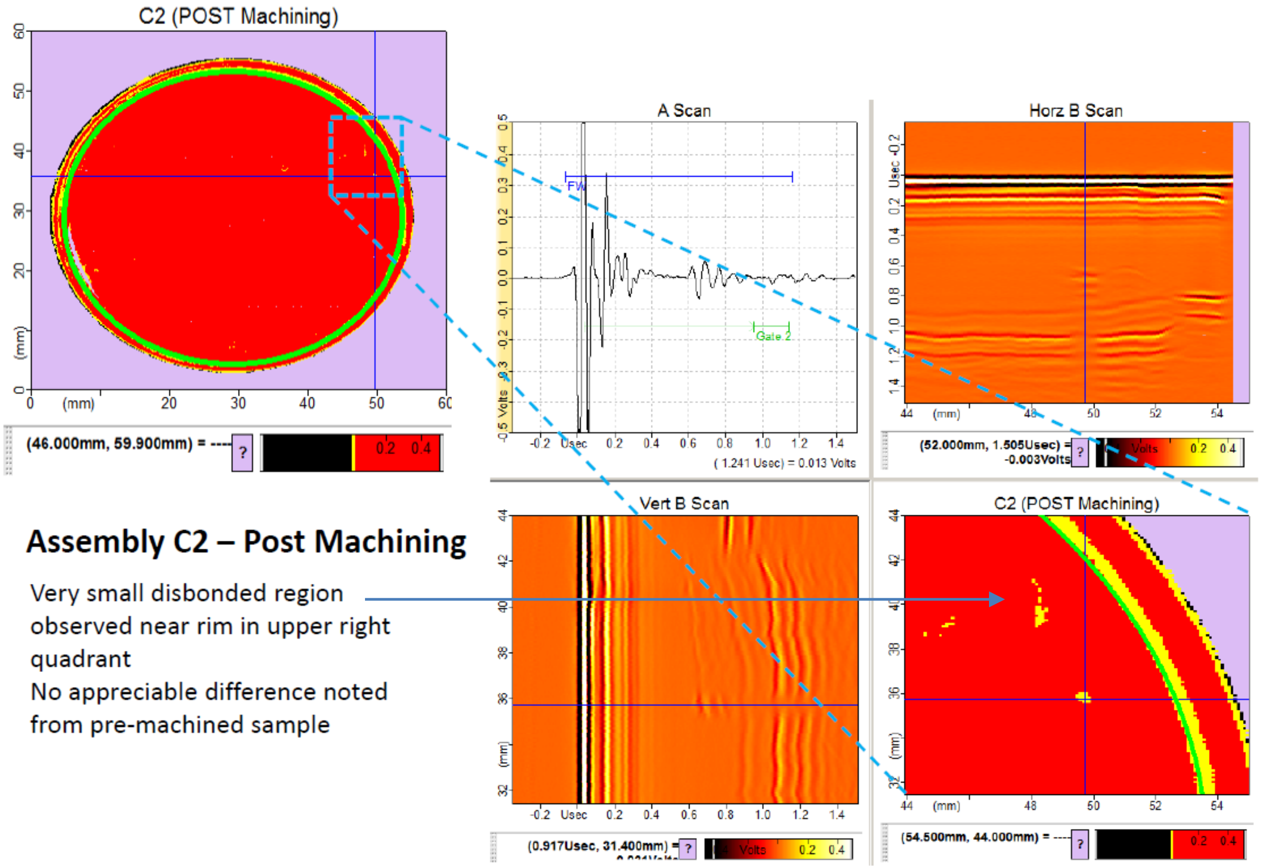
Completed thick disks

The Zr cladding was ebeam welded and then the cans were HIP bonded to create good thermal contact between the DU and Zr.

Characterization of the Zr Clad DU Disks



Optical Image of a sectioned test piece

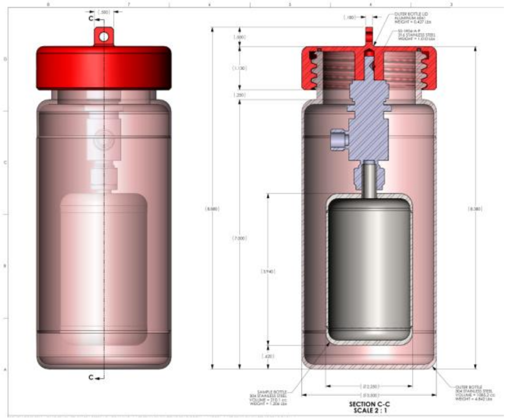


Assembly C2 – Post Machining

- Very small disbonded region observed near rim in upper right quadrant
- No appreciable difference noted from pre-machined sample

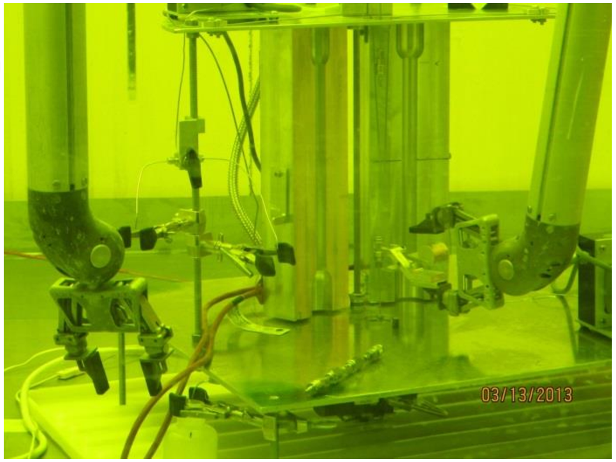
Ultrasonic test results of the final machined component.

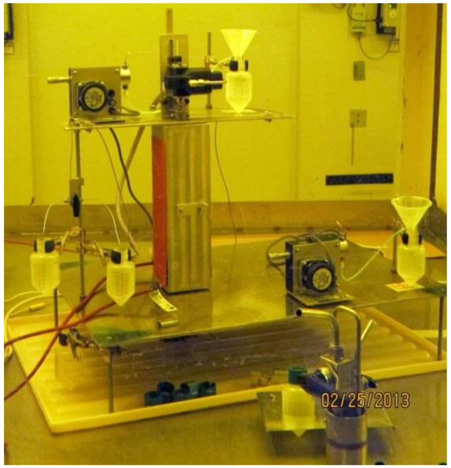
Experimental Validation
Low Enriched Uranium Irradiations
Generation of *ca.* 1 mCi ^{99}Mo



Experimental Validation
Low Enriched Uranium Irradiation
Generation of 70 mCi ^{99}Mo

Experimental Validation
Separations Chemistry





Summary

- LANL is partnering closely with NNSA and the other national laboratories to help 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 increase the production of ^{99}Mo.