Accelerator Based Domestic Production of $^{99}$Mo


Argonne National Laboratory, 9700 S. Cass Avenue, Argonne, IL 60439

2015 Mo-99 Topical Meeting
Boston, MA
August 31 – September 3, 2015
SHINE support: Argonne mini-SHINE experiment

- Argonne’s mini-SHINE experiment uses fissioning of uranyl-sulfate solutions using photo-neutron target at Argonne electron linac to produce Mo-99, this experiment is designed to:
  - Study the effects of fission on target-solution chemistry and radiolytic off-gas generation
  - Demonstrate the recovery and purification of $^{99}$Mo from an irradiated target solution
    - Produce Mo-99 to ship to potential Tc-99m generator manufacturer partners

Phase 1 (ongoing)
- Linac operates at 35 MeV and 10 kW beam power on the Ta target
- 5 L solution (140gU/L) are irradiated with neutrons generated through gamma-n reaction in tantalum target
- Maximum solution power is $\leq 0.05$ kW/L
- Up to 2 Ci of Mo-99 will be produced

Phase 2
- Experiment will be conducted at 35 MeV beam energy and up to 30 kW beam power
- 20 L solution will be irradiated with neutrons generated in a depleted-uranium (DU) target (Zr cladded DU discs were manufactured at LANL)
- Maximum solution power will be $\leq 0.5$ kW/L
- Up to 20 Ci of Mo-99 will be produced
Phase 2 mini-SHINE irradiation setup
Overview of DU Target Design

- Beam Window and Vessel Wall
- DU Disks (Typ.)
- Coolant Plenums
- Flow Channels Across Disk Faces (Typ.)
- Containment Vessel Wall
- Flow Divider and Spring Housing
- Coolant Inlet and Outlet
DU Target Design Parameters

- **Design**
  - Material for all parts of the assembly: 316 SS.
  - Heat load due to internal heat generation from the electron beam:
    - Total power of electron beam: 20 kW
    - Calculated total power absorbed by target: 16.3 kW
    - Maximum heat generation rate: 4.94 kW/cm$^3$
  - Maximum operating temperatures are:
    - 300 °C at the center of the DU disks (to prevent grain growth and clad fatigue stress)
    - 100 °C at the surface of the disks (to prevent boiling of the water coolant).
  - Maximum water coolant flow velocity: 12 m/s
  - Maximum internal pressure from water coolant: < 50 psig.

- **Handling After Irradiation**
  - Target assembly shall be removed into connecting cask and coolant lines drained and disconnected
  - Disconnect of all tubing lines shall be accomplished outside of the shielded box
  - Adequate moveable shielding shall be provided for removal of the target

- **Quality Assurance**
  - Welds shall be ASME B&PV Code certified (Note that this is not a pressure vessel).
  - Fabrication vendors shall meet ANL Procurement Level B.
DU Target Design
Exploded View Showing Disk Arrangement

- Spacer Thin
- Target Disk Thin
- Spacer Thick
- Target Disk Thick
- Support Bar and Flow Divider
- Coolant Outlet
- Coolant Flow Across Disk Face
- Flow Control Orifices
- Disk Installed in Spacer
- Compression Spring Housings and Flow Divider
- Initial Spring Force=10#
- ¼” Allowable Movement
- Final Spring Force=12#
DU Target Disk Fabrication by LANL

- An integral bond between the Zircaloy & uranium is critical to heat removal
  - Generally unbonded areas do not provide sufficient thermal conduction across the interface.
  - Based on past experience unbonded areas should not exceed
    - a single area larger than 1/16” diameter equivalent
    - total un-bonded area of more than 2%
  - However, many disks will have very low internal heat generation
    - disks that do not meet the above criteria may be used here.
    - Further, thermal hydraulic analysis may be used to qualify disks that do not meet criteria
  - Also, clad failure due to these mechanisms is slow and monitoring of the coolant water for contamination is a viable method for determining of target’s end of life.

- Large grain size in the uranium is evident in the images.
  - a result of HIP bonding at elevated temperatures that are in the beta phase of the uranium and then allowed to slow cool. (Required for good bonding).
  - Large grains may cause swelling due to irradiation and directional growth due to thermal cycling of the uranium.
  - However, required target lifetime is relatively short and operating temperatures of the uranium are low.
DU Target Thermal/Hydraulic Analysis Model

- Flow rate: 5 gpm per channel
- 2\textsuperscript{nd} disk absorbs largest amount of energy: \(~3\ kW\)
- Geometry of cooling channels modified to obtain optimum velocity across the face of the disk
DU Target Thermal/Hydraulic Analysis
Normal Operating Conditions

Disk Surface Temperature

FWHM=6mm

FWHM=7.5mm

FWHM=9.2mm

Disk Center Temperature

100 °C at Surface of Disk
FWHM = 7.5 mm

Normal Operating Conditions
95 % of power absorbed
FWHM = 9.2 mm

Disk Radius (cm)

The volumetric heat generation rates presented here assume an electron beam power of 20 kW.
DU Target Thermal/Hydraulic Analysis

Time for Temperature Increase

- Initial conditions from results of steady state analysis of normal operating conditions
- Change heat generation to FWHM = 6 mm
- Transient analysis begins at time step 100
- \(~120\) ms to reach \(100^\circ\) C at the surface of the disk
- \(~200\) ms to reach steady state
DU Target Thermal/Hydraulic Analysis
Off-Center Beam

Miss-alignment of the beam does not cause overheating of the discs

Disk Horizontal Direction (cm)

Disk Surface Temperature

Disk Center Temperature
Overview of 20L Process Tank Design

- Inner Process Tank
- Outer Cooling/Moderator Tank
- Target Sleeve Thru both Tanks
- Instrument and Rabbit Penetrations
- Connections for Top Cooling Coil and Condenser/Heat Exchanger and Process
- View Port
- Removable Flange
- Process Fluid Level
- Inner Process Tank
- Overall Tank Assembly Size: $\phi 22'' \times 22''$ H
20L Tank Thermal/Hydraulic Analysis

Temperature Contour at Center Plane

Maximum Process Fluid Temperature = 86 °C

Maximum Wall Temperature = 44 °C

Velocity vectors at center plane
Cooling System P/I Diagram
Summary

- Phase 1 mini-SHINE experiment will be finished within two months.
- Major components of the Phase 2 mini-SHINE experiments are designed and procured.
- Installation will begin as soon as Phase 1 program is finished.
- Thermal hydraulic performance of the target is evaluated. Target can dissipate 20 kW beam power.
- Off-normal scenarios for target operation were investigated. Significant deviation from the center does not lead to elevated temperatures. Significant decrease of the beam diameter can lead to surface boiling in the target, so beam size has to be monitored.
- Low density of defects and their small size for the DU cladded discs will allow a large number of thermal cycles for the target.
- CFD calculations for 20L solution tank predict maximum temperature of the solution $T=86^\circ$C and good mixing.
Proof of Concept Demonstrations for Electron Accelerator Production of $^{99}$Mo

- Under the direction of the NNSA, ANL and LANL are partnering with NorthStar Medical Isotopes, LLC. to demonstrate and develop accelerator production of $^{99}$Mo through the $^{100}$Mo($\gamma,n$)$^{99}$Mo reaction.
  - The threshold for the reaction is 9 MeV.
  - The peak cross section is 150 mb at 14.5 MeV.
- High energy photons are created with a high power electron beam through bremsstrahlung.
- Enriched $^{100}$Mo is commercially available for $400$-$600$ per gram for kg quantities.
# Production irradiations (January - September 2015)

## Production Test Matrix

<table>
<thead>
<tr>
<th></th>
<th>Thermal Test</th>
<th>Production Test 1</th>
<th>Production Test 2</th>
<th>Production Test 3</th>
<th>Production Test 4</th>
<th>Production Test 5</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Purpose</strong></td>
<td>Validate the thermal performance of the target</td>
<td>Test Enrichment 1 at high energy</td>
<td>Test Enrichment 2 at high energy</td>
<td>Test Enrichment 3 at high energy</td>
<td>Test Enrichment 2 at low energy</td>
<td>Test Enrichment 4 at high energy for long duration</td>
</tr>
<tr>
<td><strong>Energy (MeV)</strong></td>
<td>42 and 35</td>
<td>42</td>
<td>42</td>
<td>42</td>
<td>35</td>
<td>42</td>
</tr>
<tr>
<td><strong>Current (uA)</strong></td>
<td>300 and 550</td>
<td>95</td>
<td>180</td>
<td>180</td>
<td>222</td>
<td>180</td>
</tr>
<tr>
<td><strong>Power (kW)</strong></td>
<td>12.6 and 19.3</td>
<td>3.5</td>
<td>7.5</td>
<td>7.5</td>
<td>7.8</td>
<td>7.5</td>
</tr>
<tr>
<td><strong>Duration (hours)</strong></td>
<td>2</td>
<td>19</td>
<td>21</td>
<td>19</td>
<td>24.4</td>
<td>156</td>
</tr>
<tr>
<td><strong>Targets</strong></td>
<td>Natural</td>
<td>E1 (97.39%) and Natural</td>
<td>E2 (99.03%) and Natural</td>
<td>E3 (95.08%) and Natural</td>
<td>E2 (99.03%) and Natural</td>
<td>E4 (95.08%) and Natural</td>
</tr>
<tr>
<td><strong>Mo-99 EOB Activity [Ci]</strong></td>
<td>0.2 and 0.28</td>
<td>0.92</td>
<td>2.9</td>
<td>2.2</td>
<td>4.2</td>
<td>15</td>
</tr>
<tr>
<td><strong>Target Thermocouples</strong></td>
<td>Yes</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
</tr>
</tbody>
</table>
Cameras’ radiation stability

Camera configuration and dosimeters placement

X-ray doses

Neutron dosimeter activation
Shielding for production facility using 270 degree magnet configuration

- Dose rates have been estimated for selected locations in a proposed production facility based on the $^{100}\text{Mo}(\gamma,n)^{99}\text{Mo}$ reaction using bremsstrahlung photons produced by an electron linear accelerator.
- Dose rates were 1 mrem/hr for areas accessible by radiation workers (the service area above the target and accelerator bays adjacent to operating accelerators) and 50 rem/hr for areas accessible by non-radiation workers (the hallway outside the accelerator bays and areas external to the facility).
High power beam stop and collimator

Power deposition from 42 MeV electron beam in aluminum

High power beam stop

120kW Beam Impingment on an Aluminum Plate w/ Boiling Constraint

Target collimator
Summary

- We have conducted four production tests using enriched Mo-100 targets.
- Irradiated material was shipped to NorthStar for consecutive separation runs.
- MCNPX calculation for production-facility shielding showed that 30 cm of lead and 250 cm of concrete will be sufficient for effective shielding both neutrons and photons.
- High power beam stop and collimators capable to handle 120 kW beam power were developed.
- Camera performance during production runs suggests high susceptibility of the IR camera to radiation damage (presumably neutrons).
Acknowledgements

• This work is done in collaboration with LANL and ORNL
• The submitted manuscript has been created by UChicago Argonne, LLC, Operator of Argonne National Laboratory (“Argonne”). Argonne, a U.S. Department of Energy Office of Science laboratory, is operated under Contract No. DE-AC02-06CH11357. The U.S. Government retains for itself, and others acting on its behalf, a paid-up nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publicly and display publicly, by or on behalf of the Government.
• Work supported by the U.S. Department of Energy, National Nuclear Security Administration's (NNSA's) Office of Defense Nuclear Nonproliferation, under Contract DE-AC02-06CH11357.