Accelerator Based Domestic Production of ^{99}Mo

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SHINE support: Argonne mini-SHINE experiment

- Argonne's mini-SHINE experiment use 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

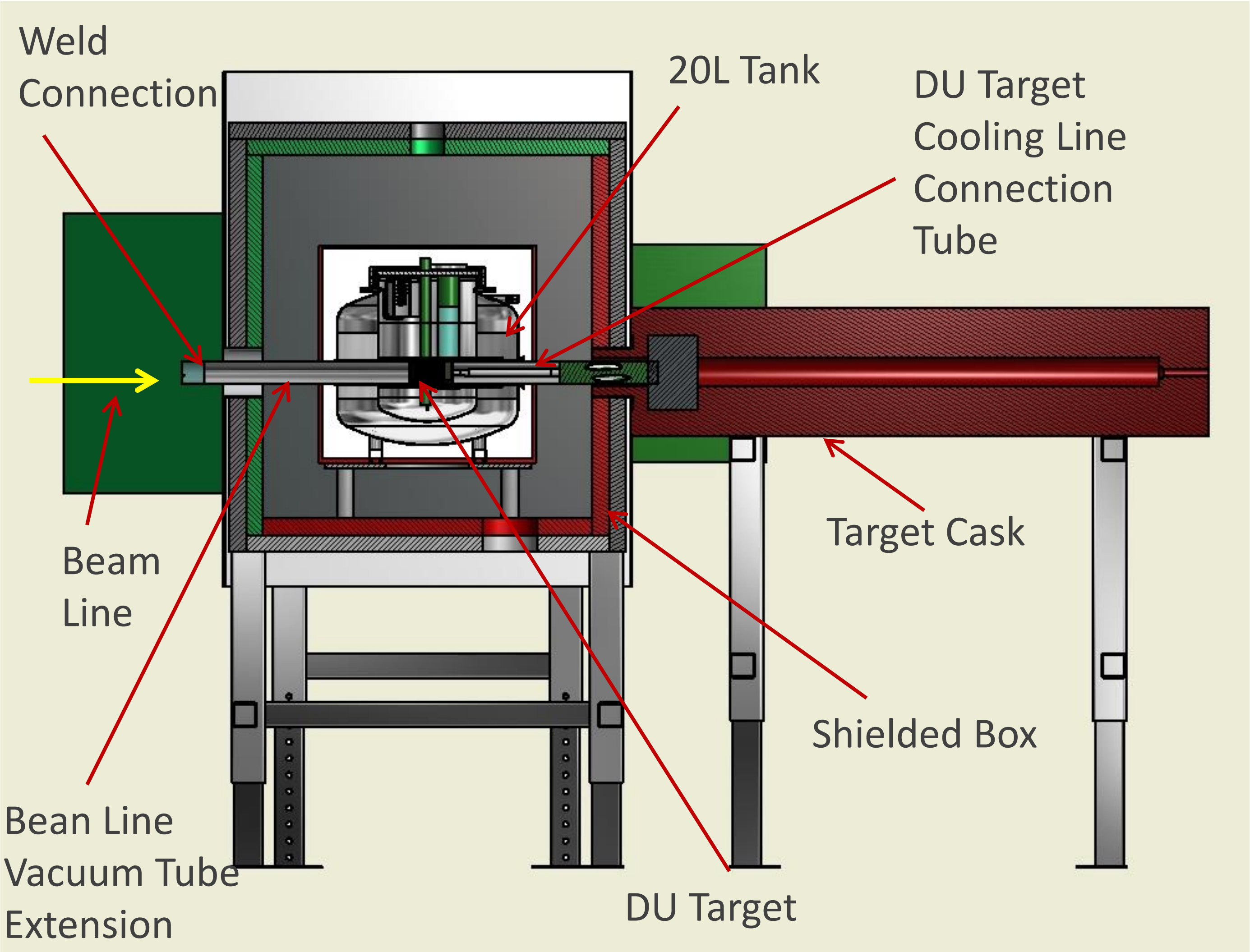
Phase 1 (ongoing)

- 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

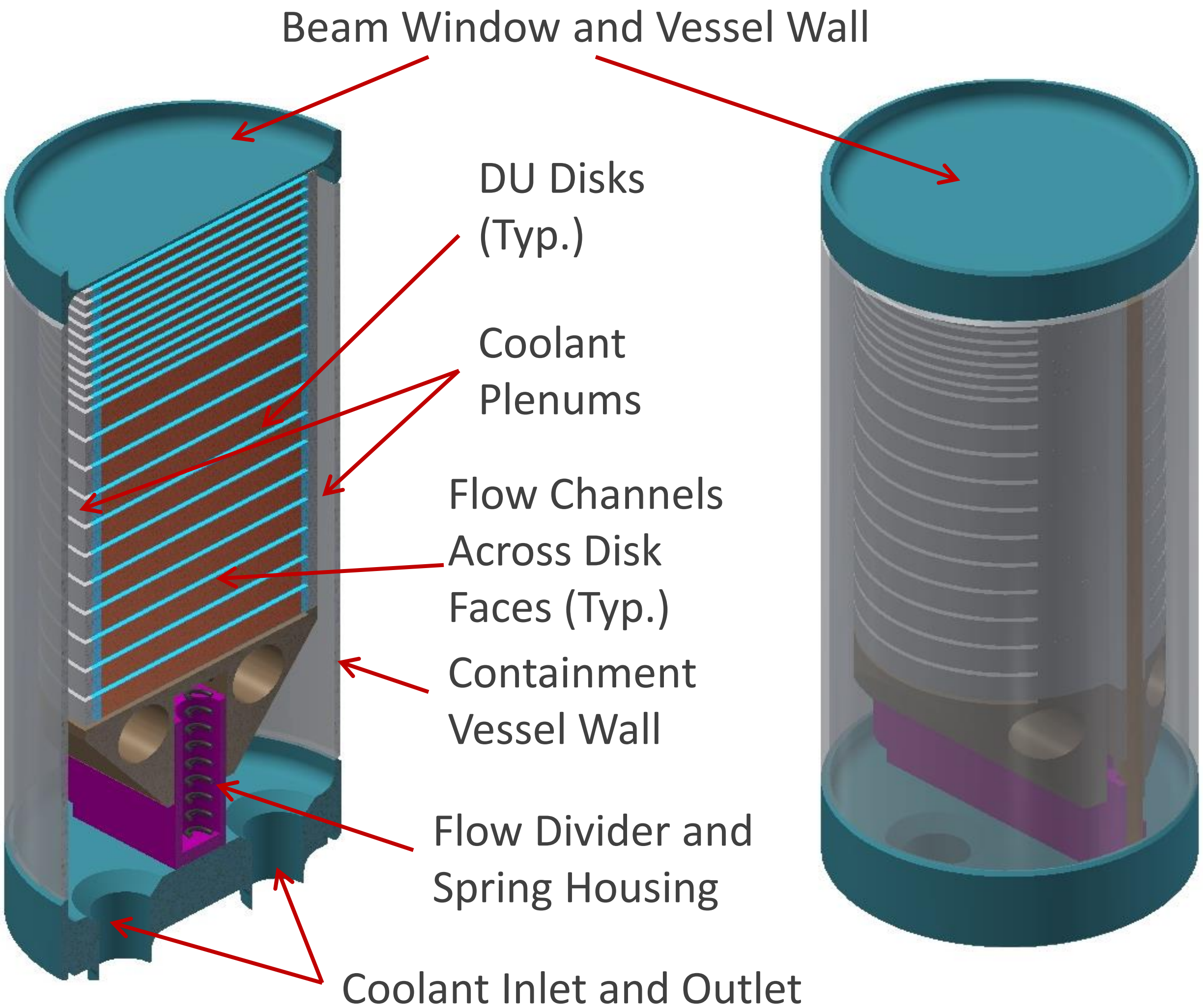
Phase 2

- 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



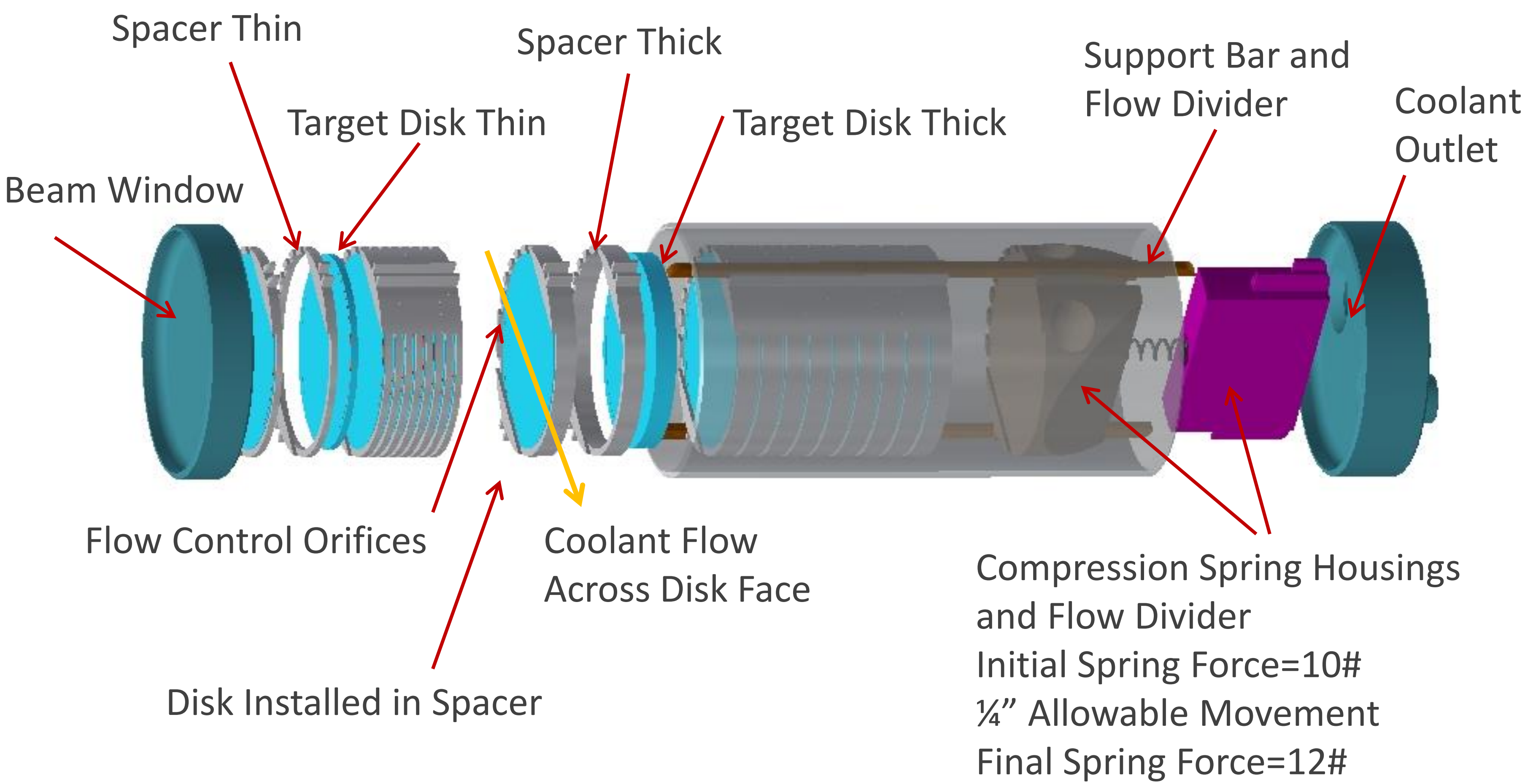
Overview of DU Target Design



DU Target Design Parameters

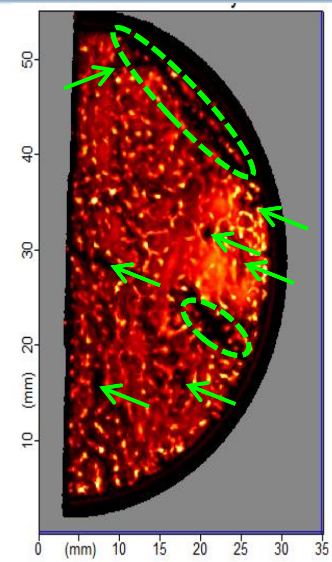
- Design
  - Material for all parts of the assembly: 316 SS.
  - Heat load due to internal heat generation from the electron beam:
    - Total power of electron beam: 20 kW
    - Calculated total power absorbed by target: 16.3 kW
    - Maximum heat generation rate: 4.94 kW/cm^3
  - Maximum operating 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.

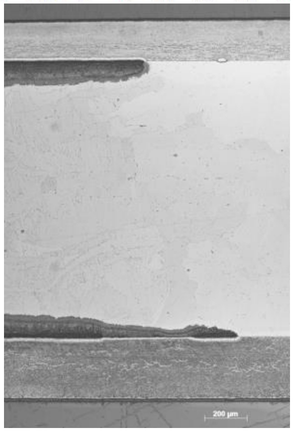
DU Target Design
Exploded View Showing Disk Arrangement

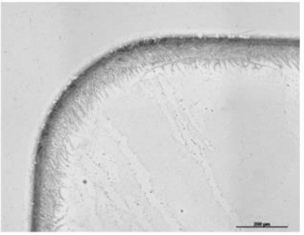


DU Target Disk Fabrication by LANL

- 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.

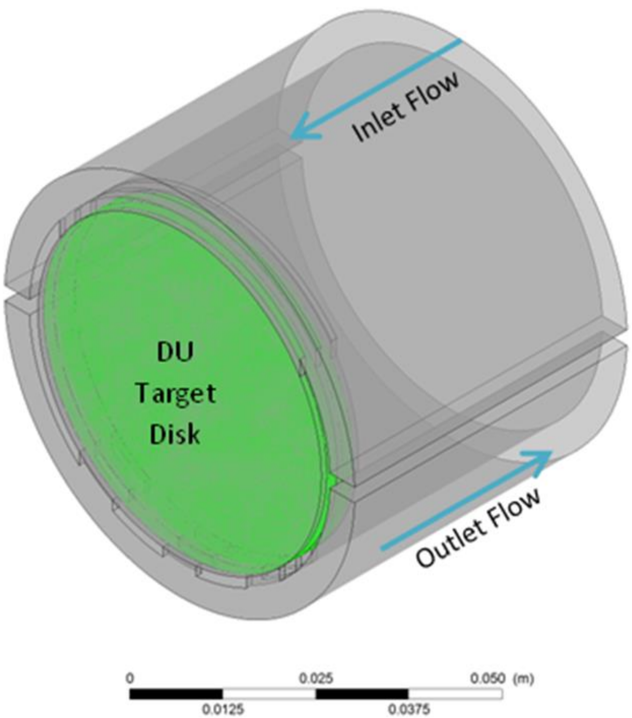


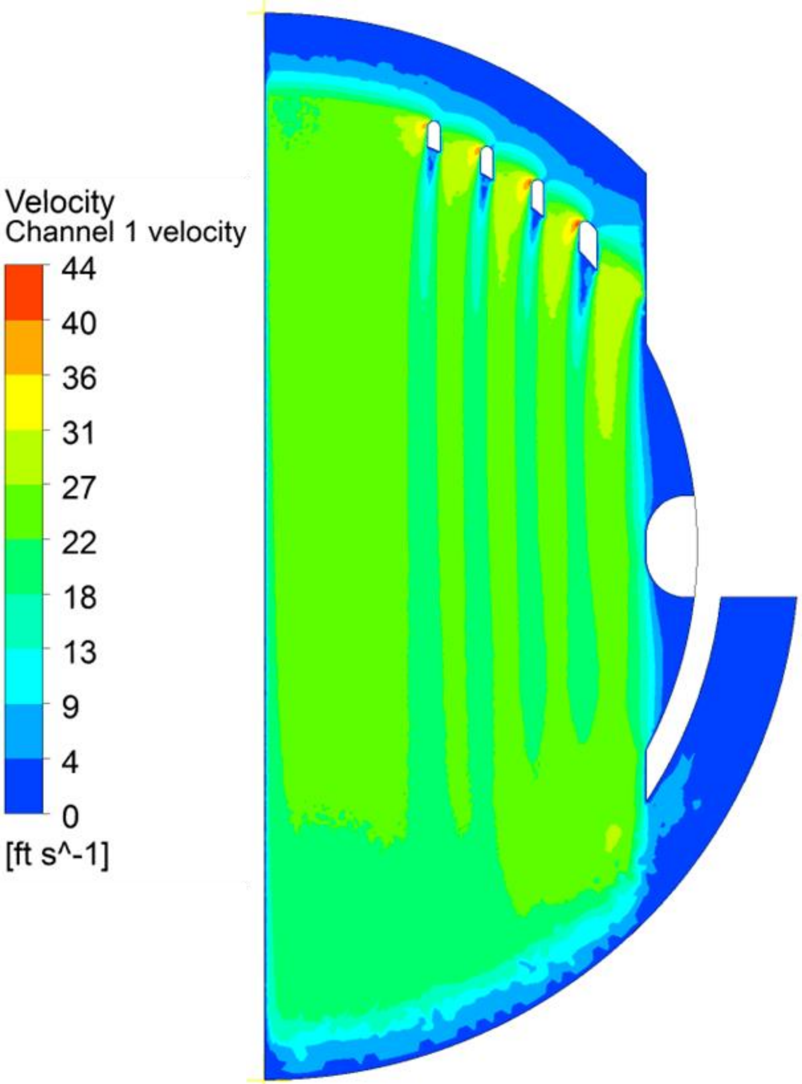




DU Target Thermal/Hydraulic Analysis Model

- Flow rate: 5 gpm per channel
- 2nd disk absorbs largest amount of energy : ~3 kW
- Geometry of cooling channels modified to obtain optimum velocity across the face of the disk

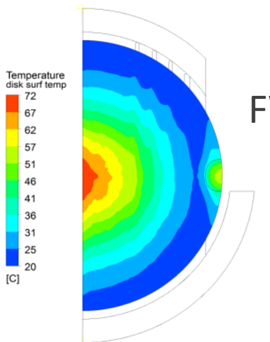




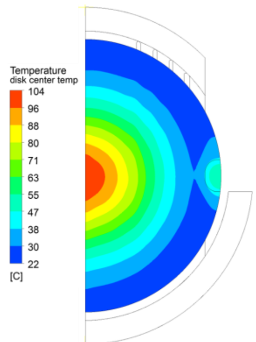
DU Target Thermal/Hydraulic Analysis Normal Operating Conditions

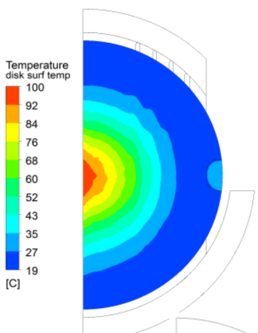
Disk Surface Temperature

Disk Center Temperature

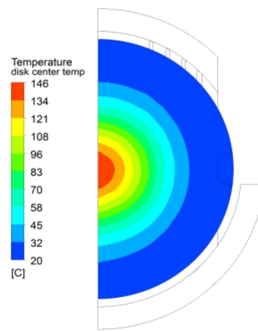


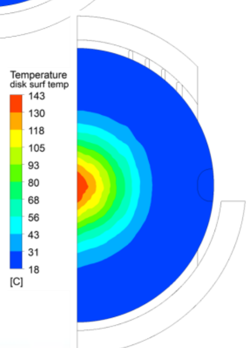
FWHM=6mm



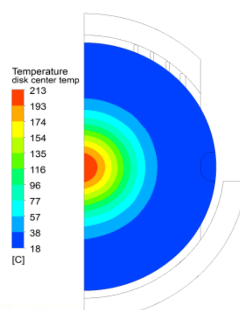


FWHM=7.5mm





FWHM=9.2mm

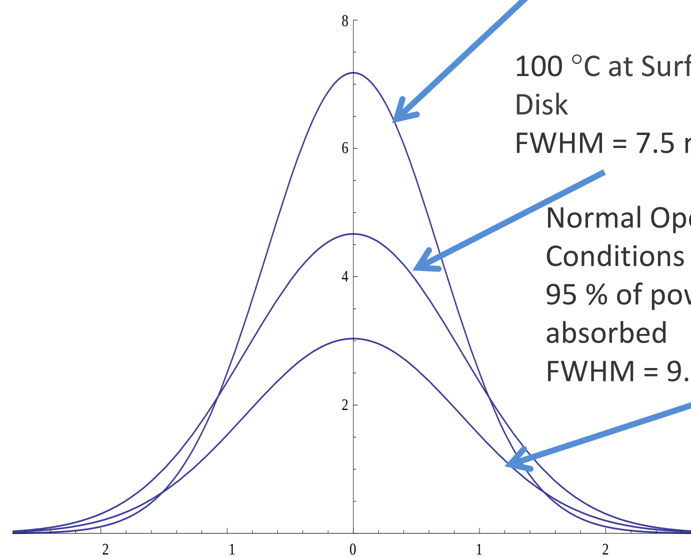


kW/cm^3

Accident Case
FWHM = 6 mm

100 °C at Surface of Disk
FWHM = 7.5 mm

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



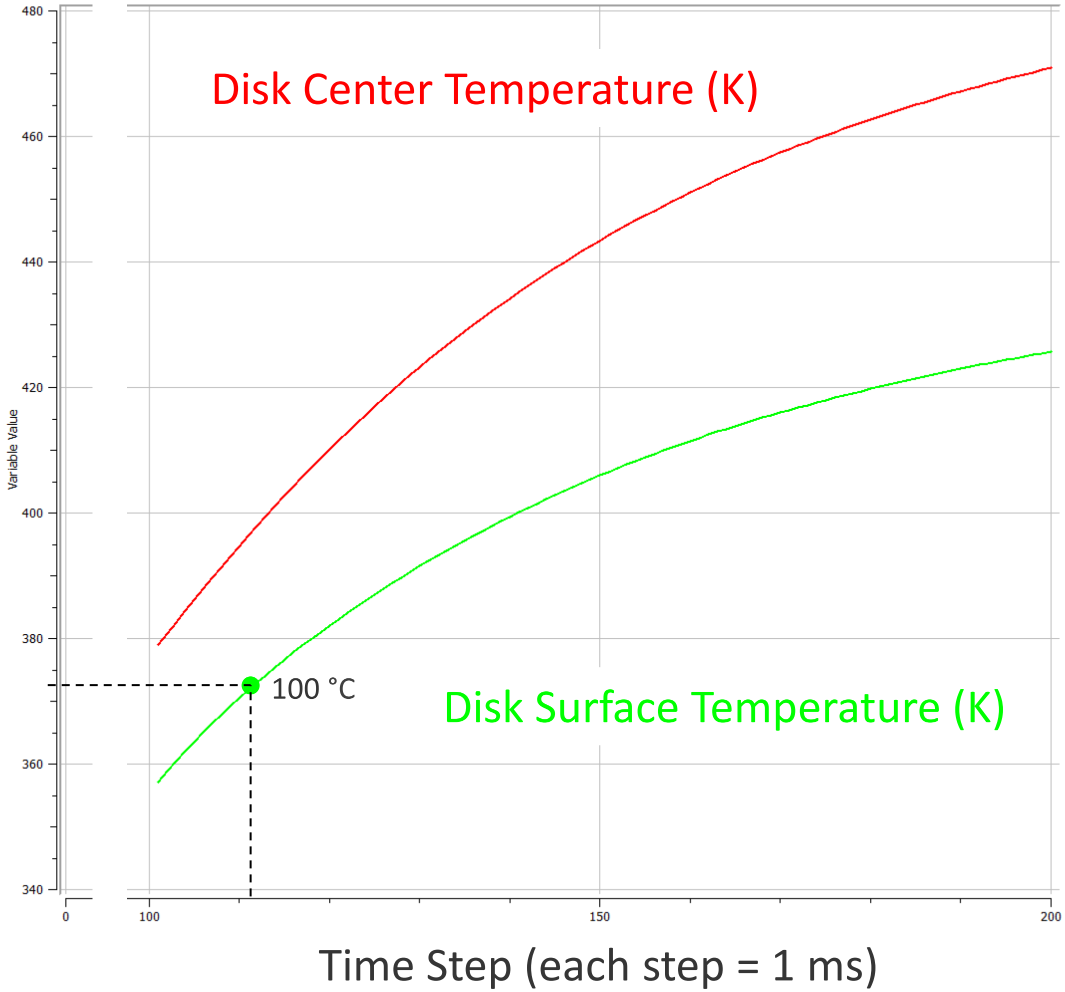
Disk Radius (cm)

The volumetric heat generation rates presented here assume an electron beam power of 20 kW

DU Target Thermal/Hydraulic Analysis
Time for Temperature Increase

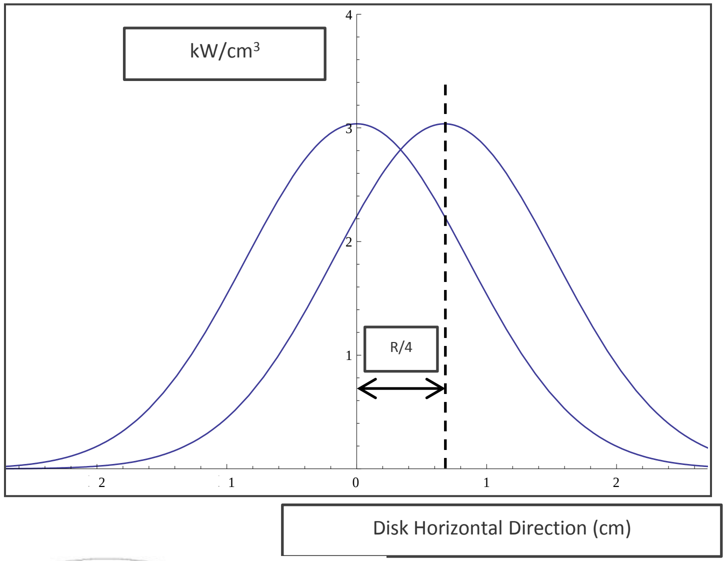
- Initial conditions from results of steady state analysis of normal operating conditions
- Change heat generation to FWHM = 6 mm
- Transient analysis begins at time step 100

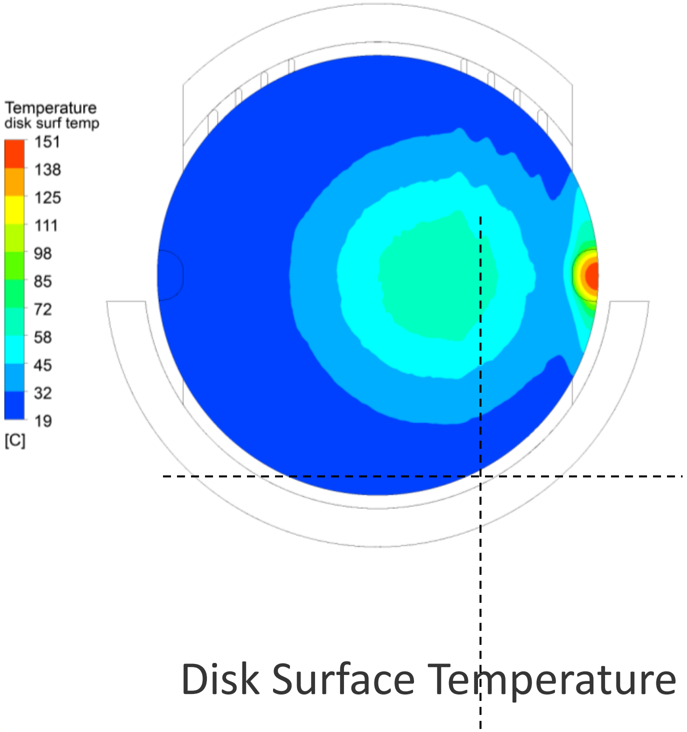
- ~120 ms to reach 100 ° C at the surface of the disk
- ~200 ms to reach steady state



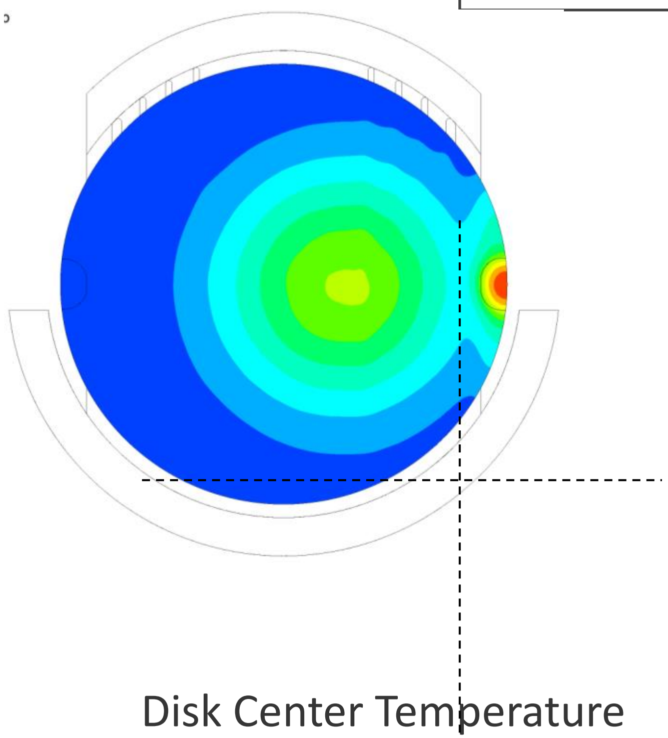
DU Target Thermal/Hydraulic Analysis
Off-Center Beam

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



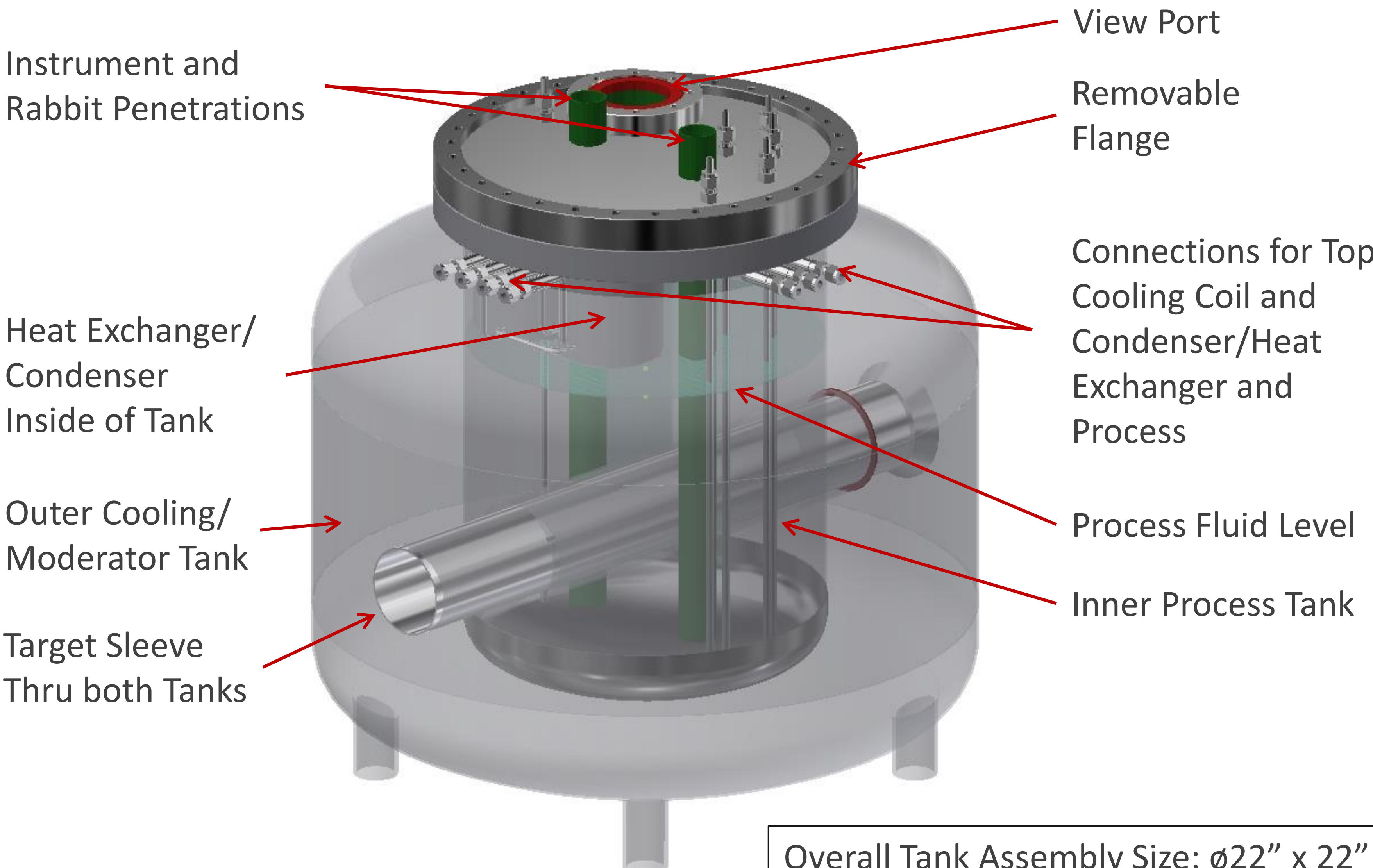


Disk Surface Temperature



Disk Center Temperature

Overview of 20L Process Tank Design

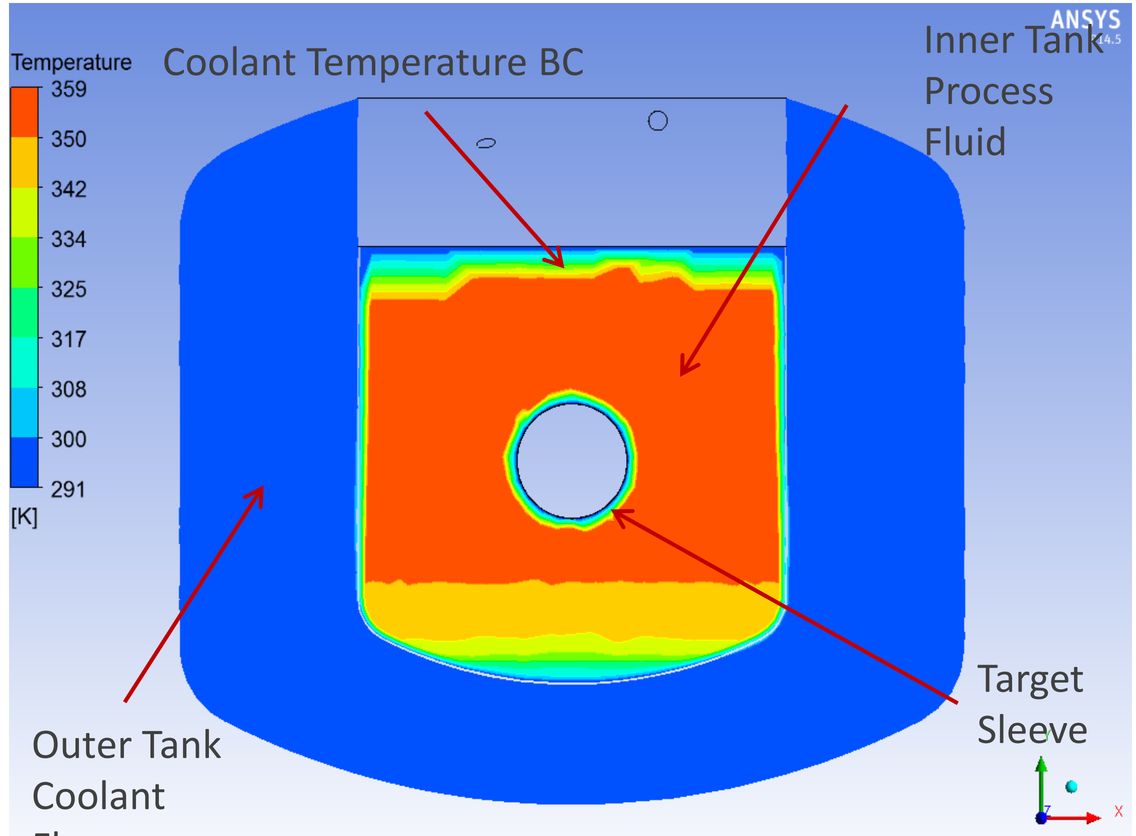


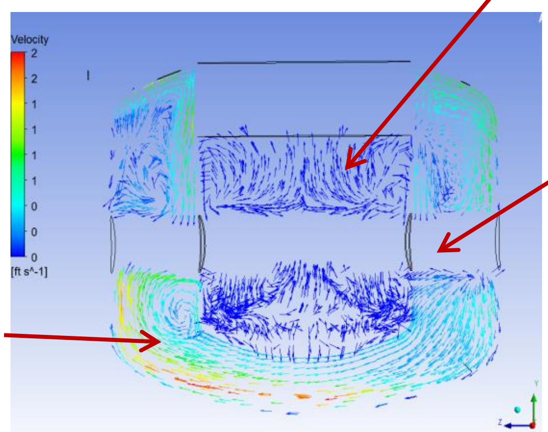
Overall Tank Assembly Size: ø22” x 22” H

20L Tank Thermal/Hydraulic Analysis
Temperature Contour at Center Plane

Maximum Process Fluid Temperature = 86 C

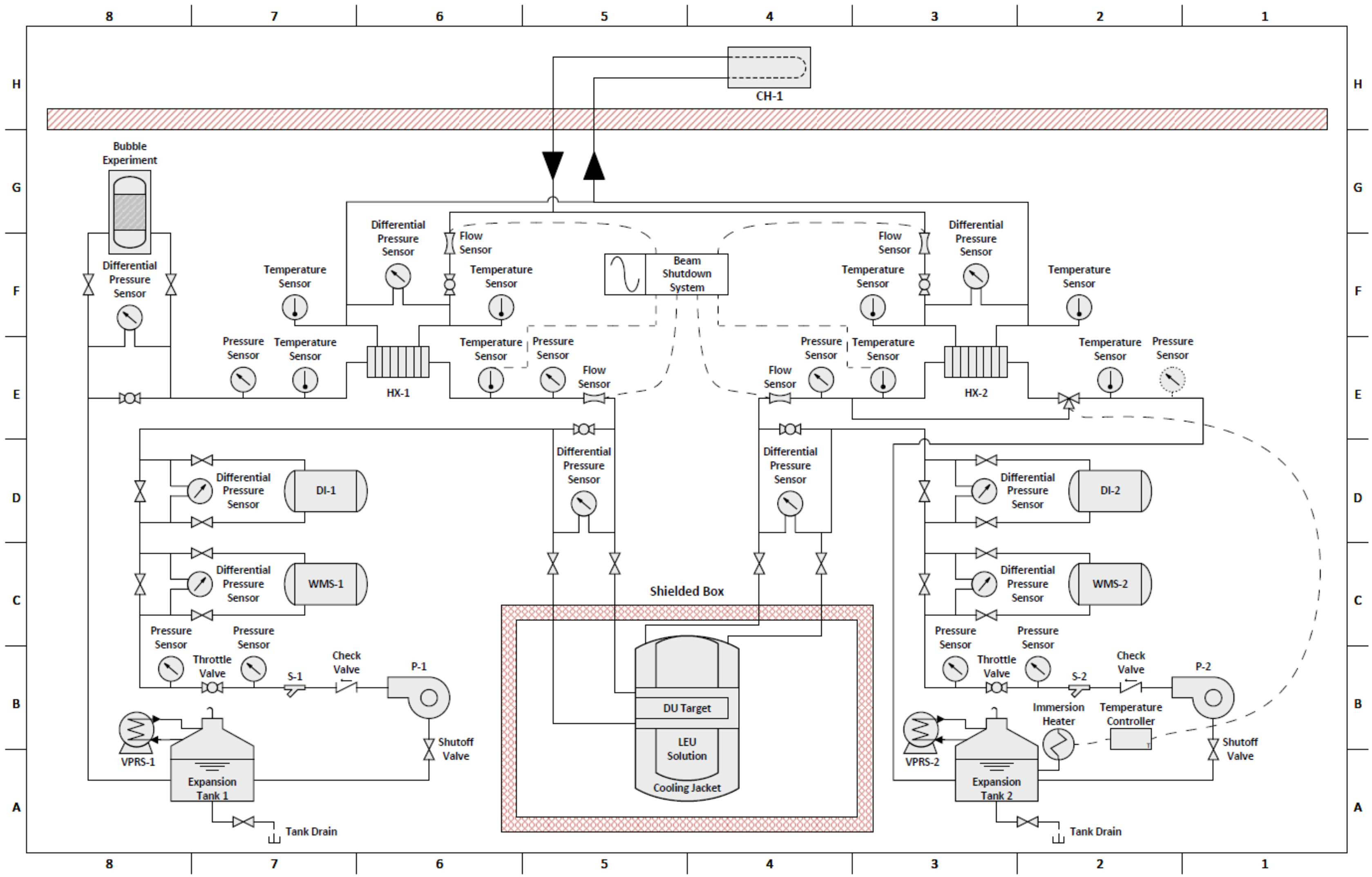
Maximum Wall Temperature = 44 C





Velocity vectors at center plane

Cooling System P/I Diagram

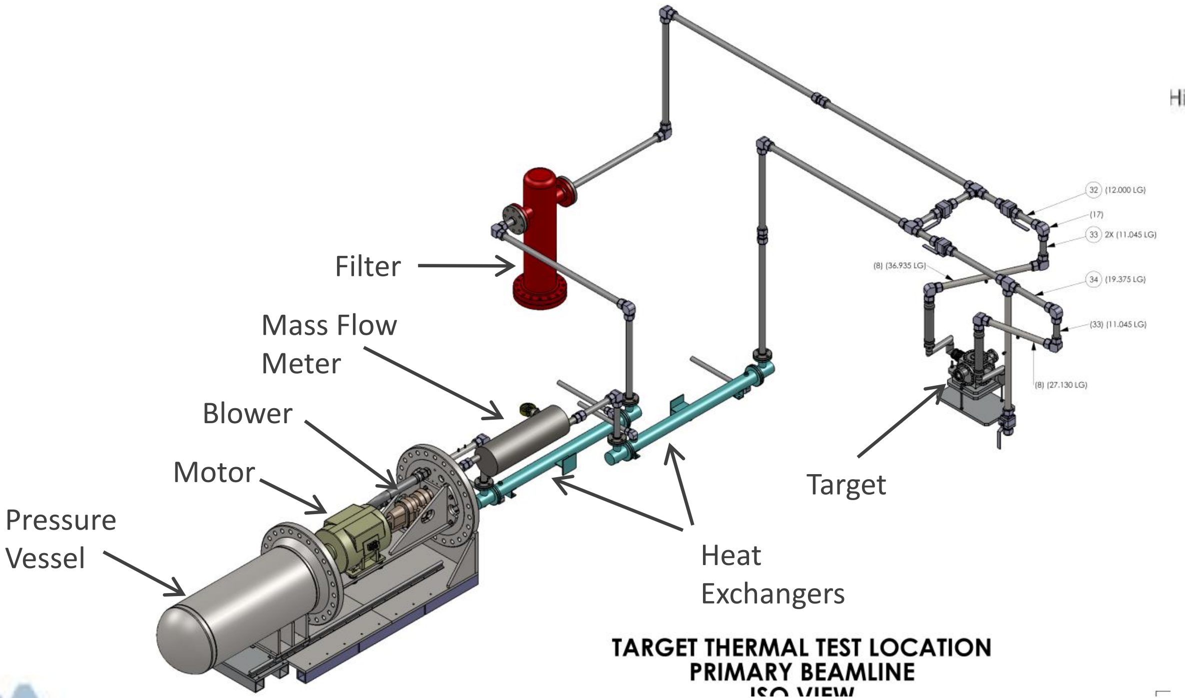


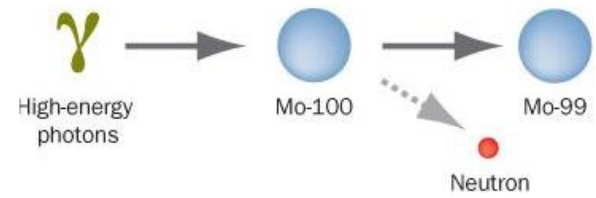
Summary

- Phase 1 mini-SHINE experiment will be finished within two month
- 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 form 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 there small size for the DU cladded discs will allow large number of thermal cycles for the target.
- CFD calculations for 20L solution tank predict maximum temperature of the solution T=86^{o}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.



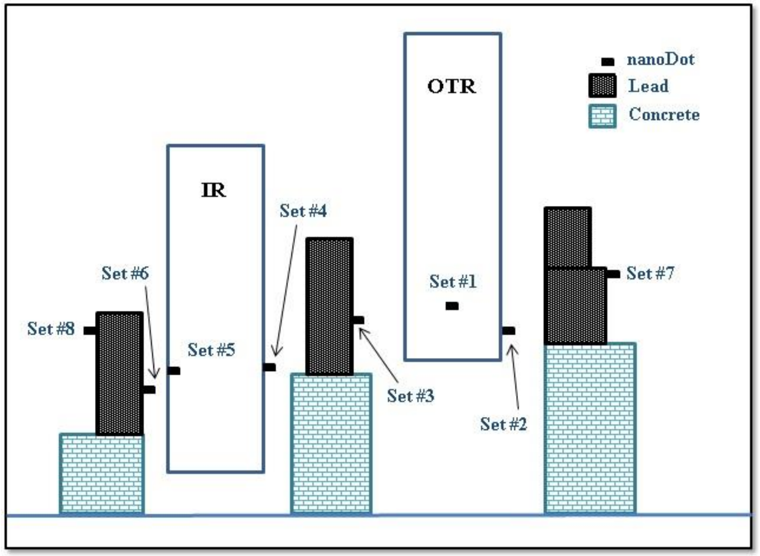


Production irradiations (January - September 2015)

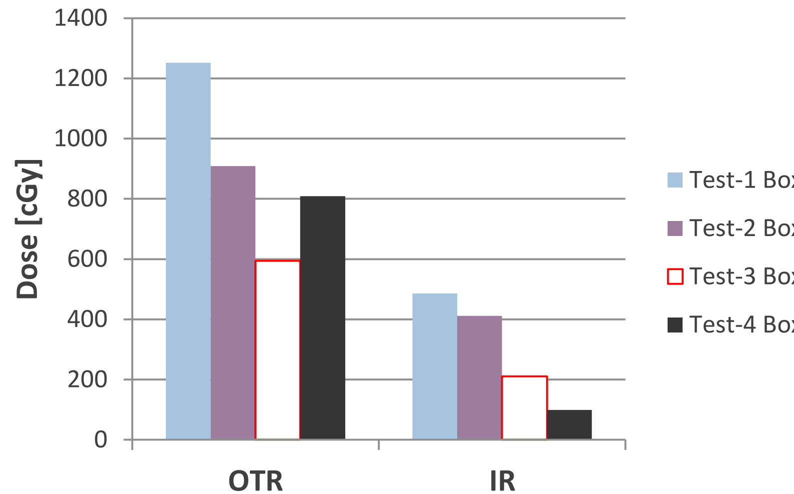
Production Test Matrix

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

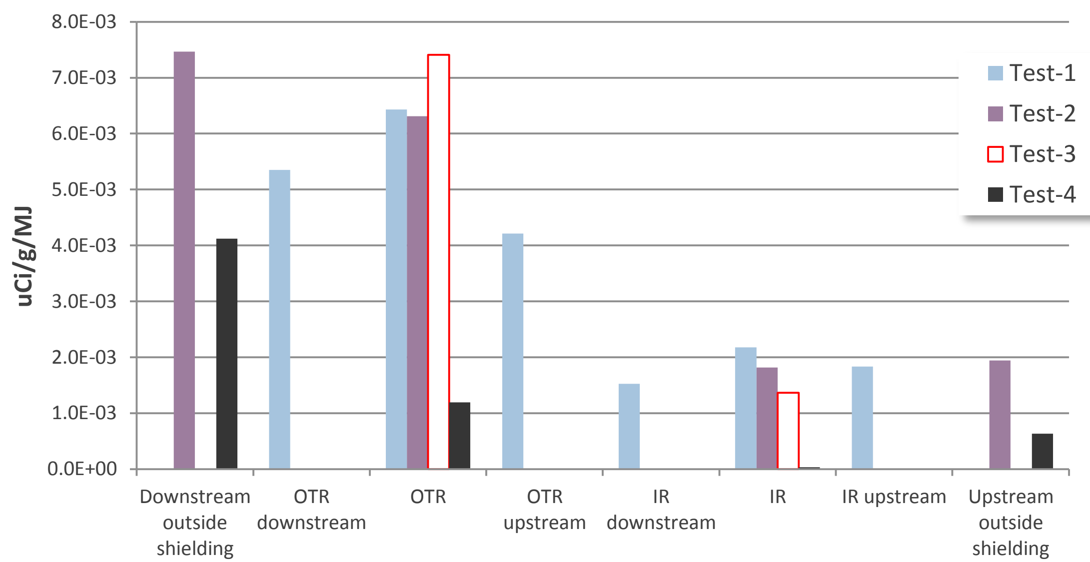
Cameras' radiation stability



Camera configuration and dosimeters placement

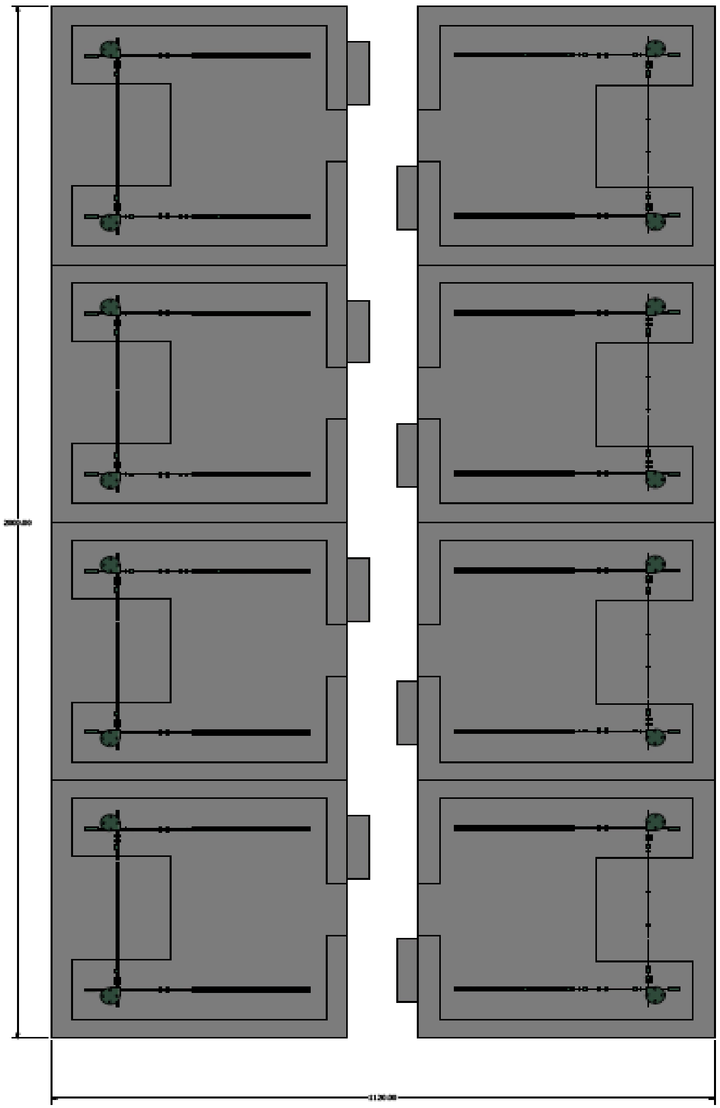


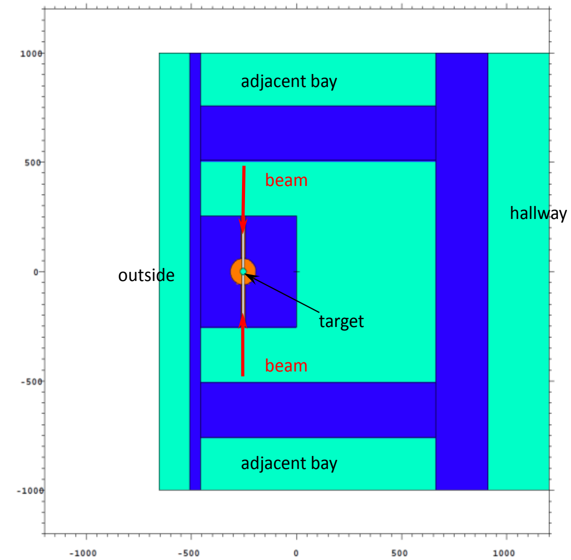
X-ray doses



Neutron dosimeter activation

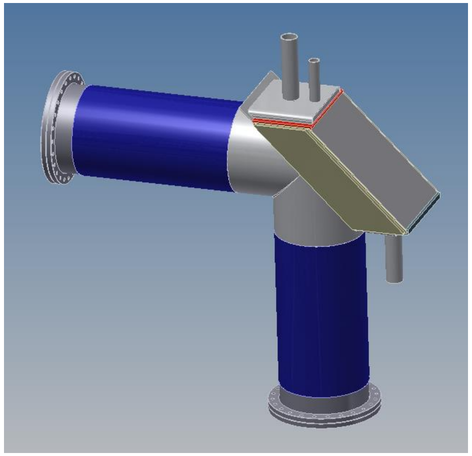
Shielding for production facility using 270 degree magnet configuration

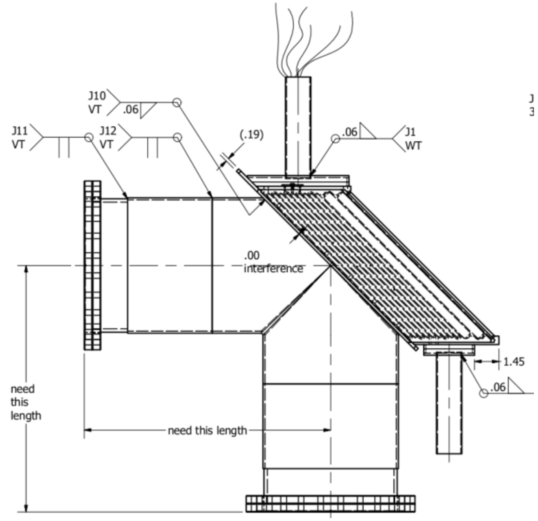


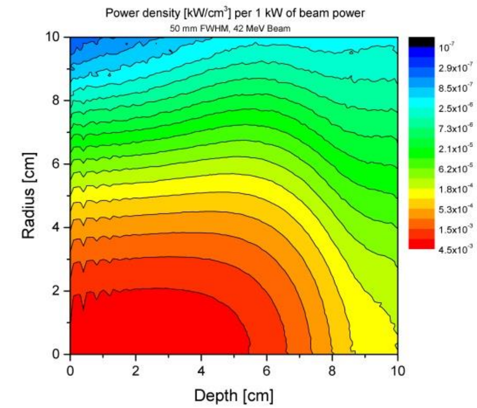


- Dose rates have been estimated for selected locations in a proposed production facility based on the $^{100}Mo(\gamma,n)^{99}Mo$ reaction using bremsstrahlung photons produced by an electron linear accelerator.
- Dose rates were 1 mrem/hr for areas accessible by radiation workers (the service area 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

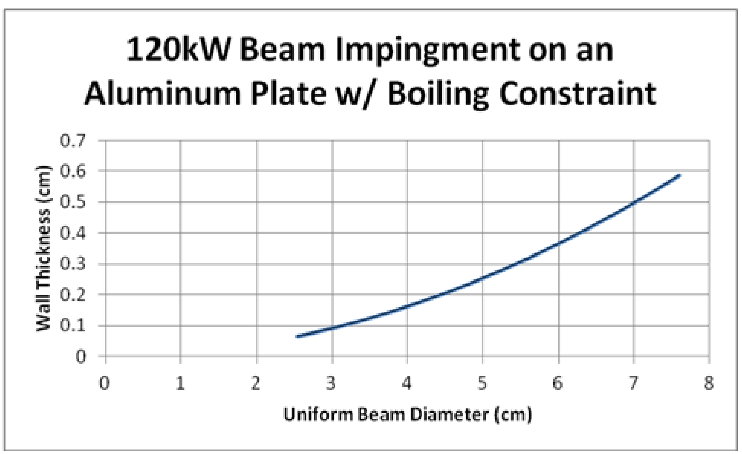


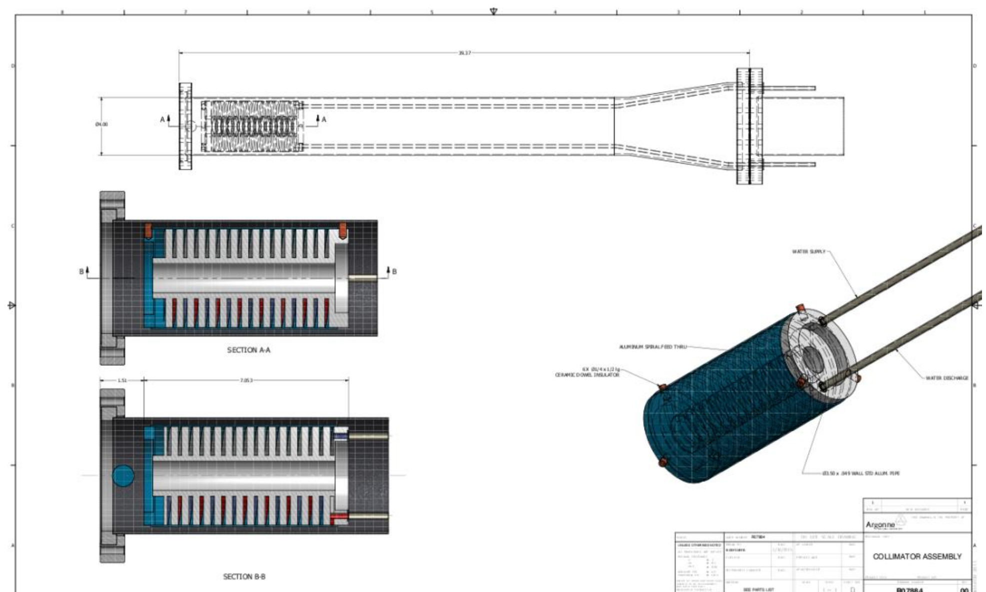




Power deposition from 42 MeV electron beam in aluminum

High power beam stop





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).

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