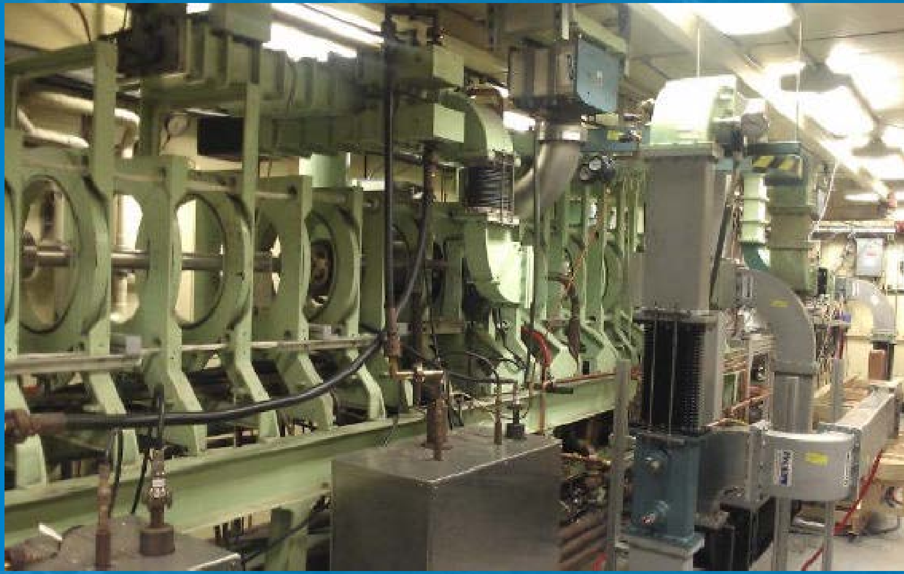
SEPTEMBER 13, 2017

DEVELOPMENT OF ACCELERATOR-BASED PRODUCTION OF MO-99



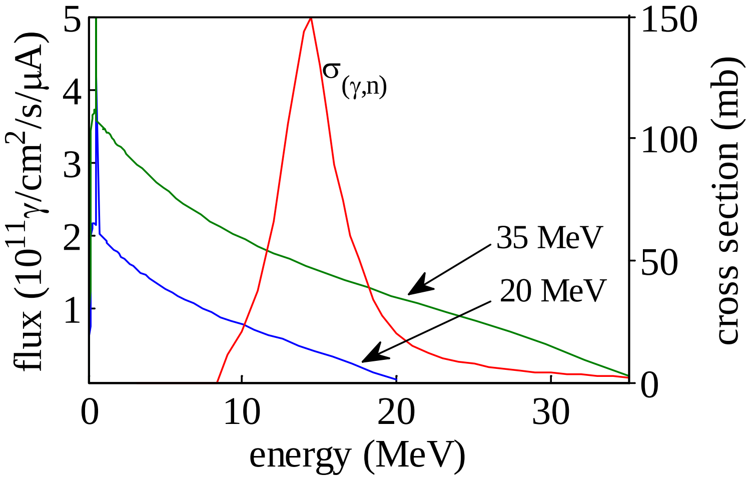
Sergey Chemerisov, Peter Tkac, Mandy Youker, Mike Kalensky, Roman Gromov, Chuck Jonah, Vakho Makarashvili, Brad Micklich, Kurt Alford, Ken Wesolowski, Kevin Quigley, Jim Bailey, Tom Brossard, David Rotsch, Jim Byrnes, and George Vandergrift

DEVELOPING A DOMESTIC SUPPLY OF Mo-99

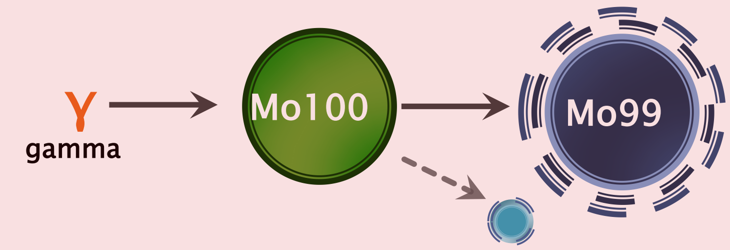
- NNSA's Material Management and Minimization Program (M^3) is assisting in the development of different technologies for producing Mo-99
- Argonne currently has development activities in those three technologies
  - Neutron activation of Mo-98
    - **NorthStar Medical Technologies, LLC**
  - Accelerator γ/n reaction on Mo-100
    - **NorthStar Medical Technologies, LLC**
  - Accelerator-driven production of fission-product Mo-99
    - **SHINE Medical Technologies**
- All technologies assert they can produce 3000 6-day Ci/week (50% of US requirements)
- This presentation will cover accelerator-related aspects of the projects

PRODUCING MO-99 WITHOUT USE OF URANIUM (NORTHSTAR)

- Under the direction of the NNSA, Argonne, ORNL and LANL are partnering with NorthStar Medical Isotopes, LLC. to develop and demonstrate accelerator production of ^{99}Mo through the ^{100}Mo(γ,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.



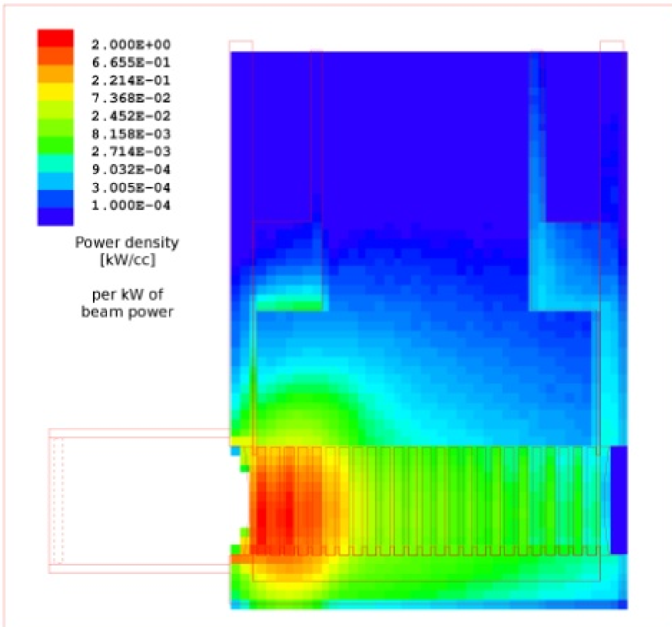
Accelerator production



Major accelerator-related tasks

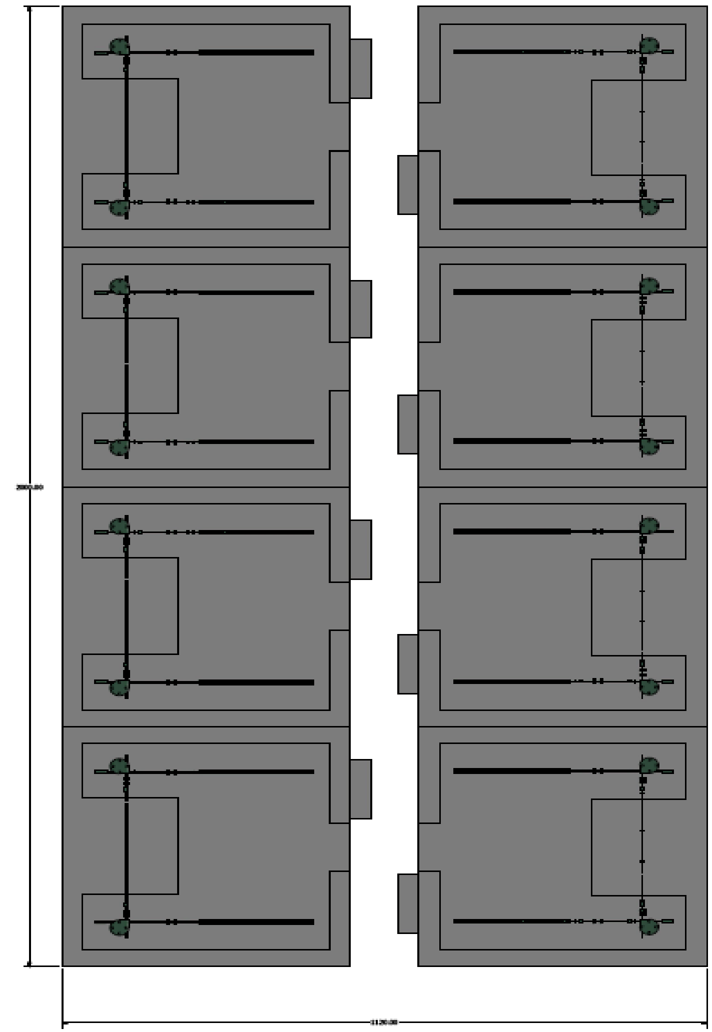
- Development and testing of beamline components
  - Achromatic bend
  - High power beam dump
  - Fast acting beam valve testing
- Target temperature monitoring
- Target window and holder material optimization
- Irradiation of sintered Mo targets
- Side reactions study

PRODUCTION FACILITY CONFIGURATION



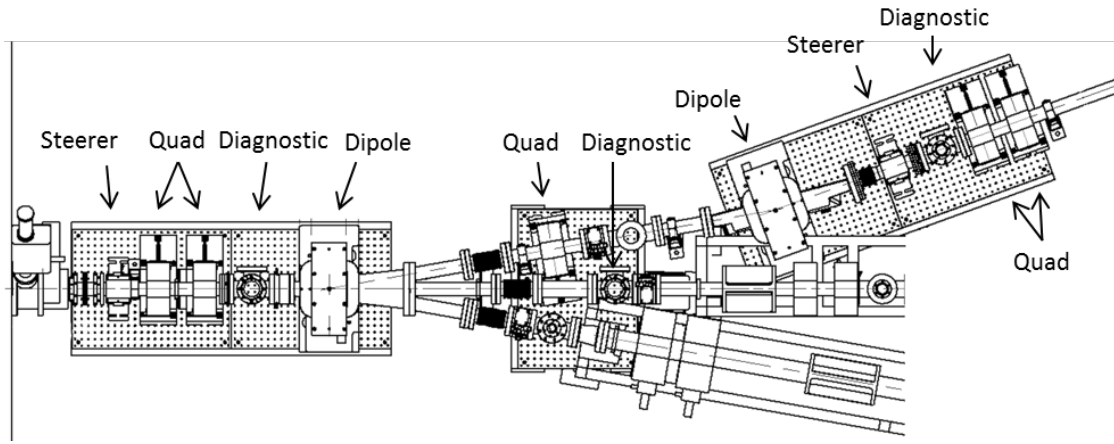
Heat deposition on the target is significantly forward peaking; irradiation from two sides will allow reduce enriched material inventory by factor of 2

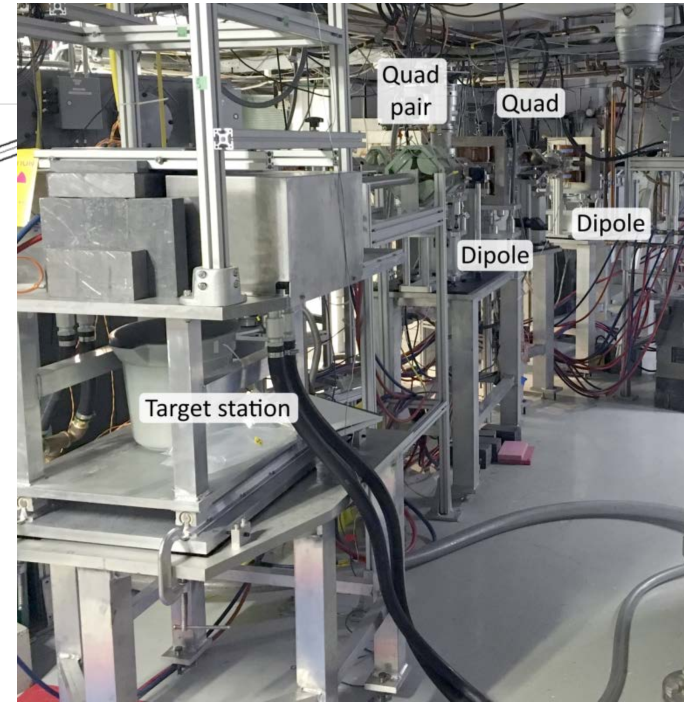
Use of two accelerators creates line-of-sight problem

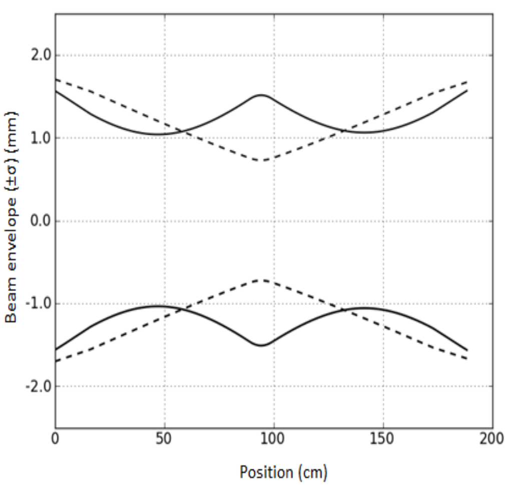


One of the possible configuration of the production facility accelerator hall

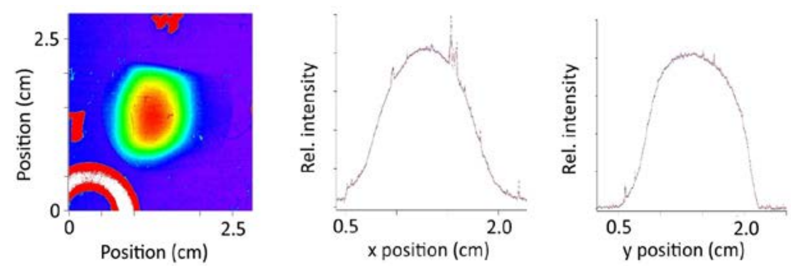
DEVELOPMENT OF ACHROMATIC BEAM BENDING SYSTEMS





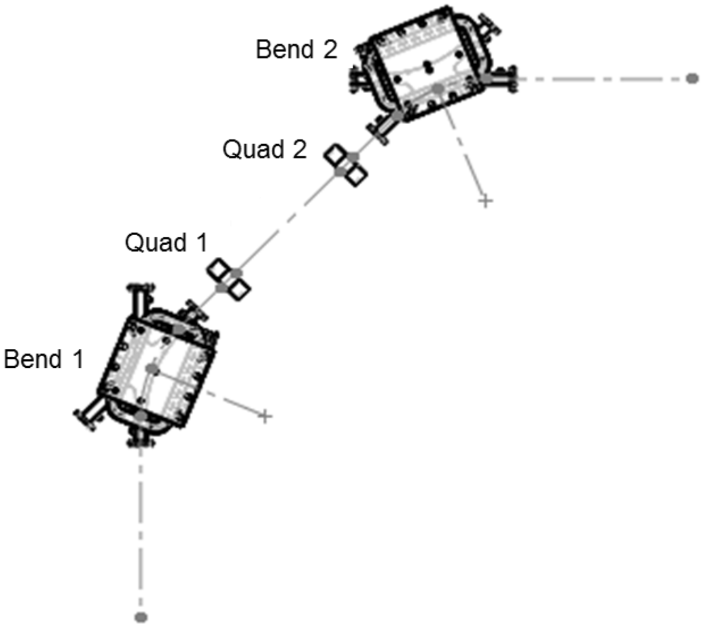


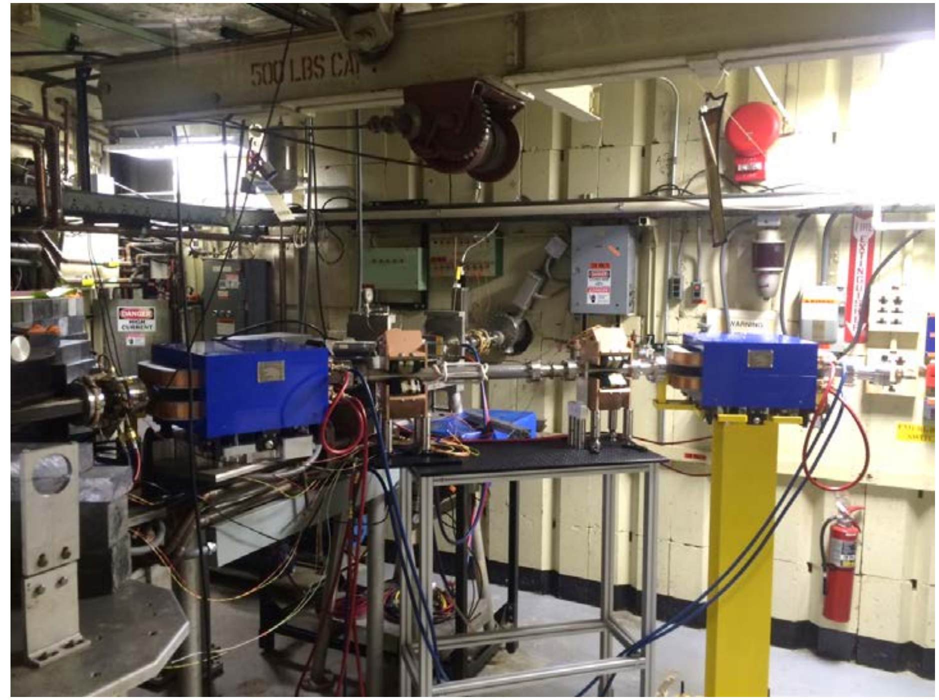
Simulated horizontal (solid) and vertical (dashed) beam envelopes for the achromatic bend assuming an energy spread of ±2%



Beam distribution on the target

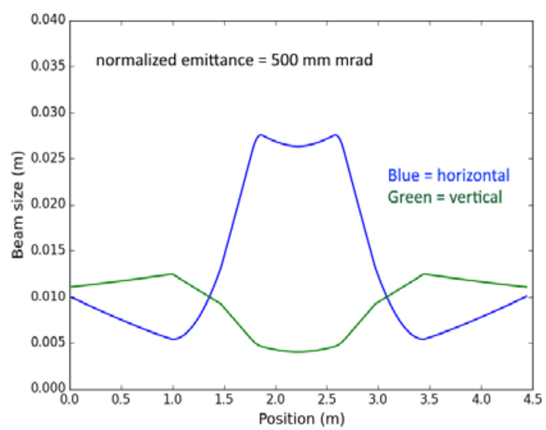
90^0 BENDING SYSTEM DESIGN AND TESTING

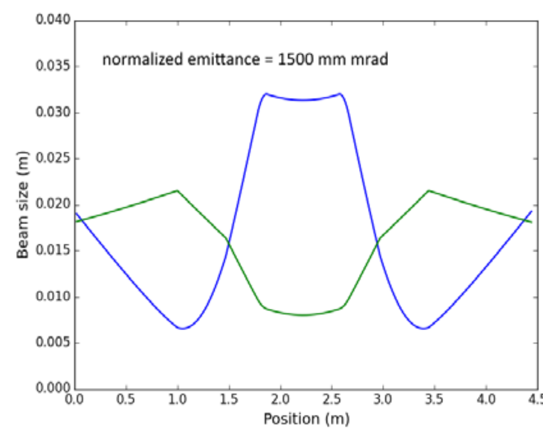




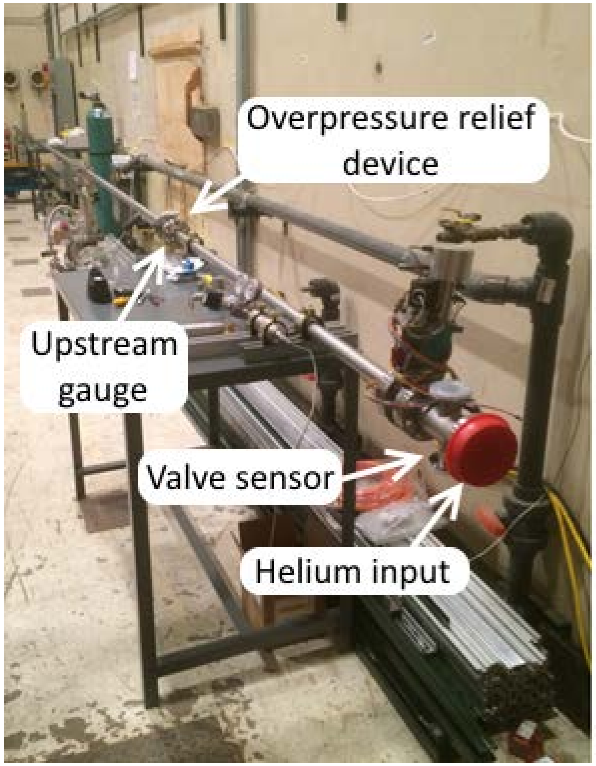
Achromatic effect would be received by using of two quadrupoles between main bends for compensation of beam dispersion.

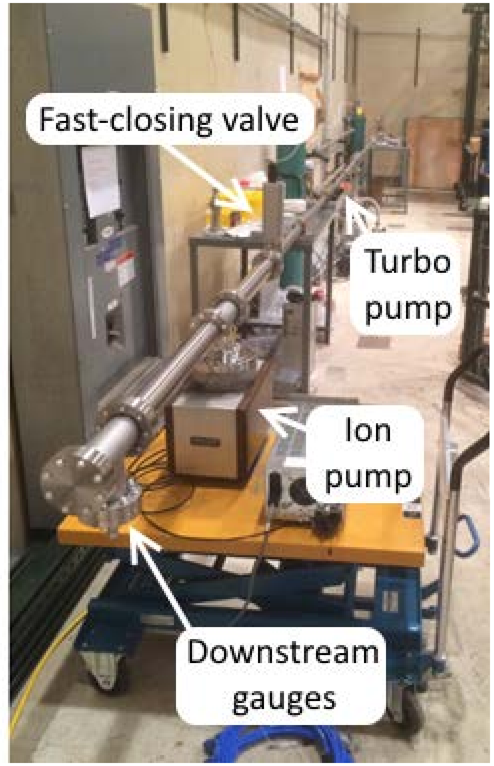
Simulations were performed with an electron beam with normalized emittance of 500 mm*mrad and 1500 mm*mrad and an energy spread of ±2.5%. First order matched beam envelopes are shown on plots.

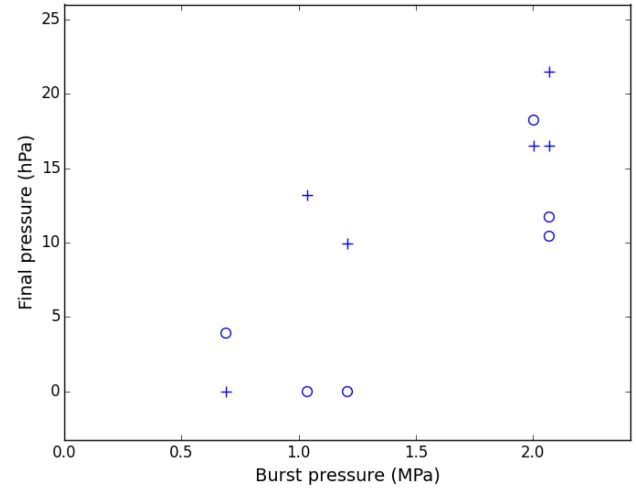


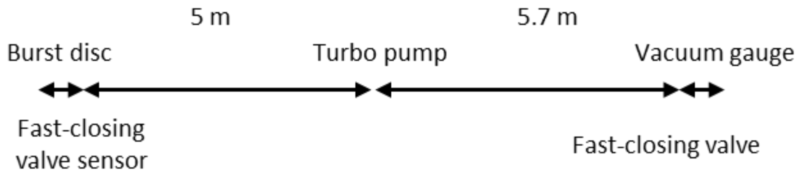


TESTING OF FAST-ACTING GATE VALVE SYSTEM







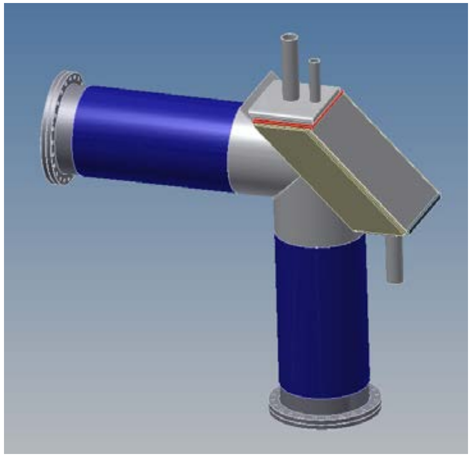


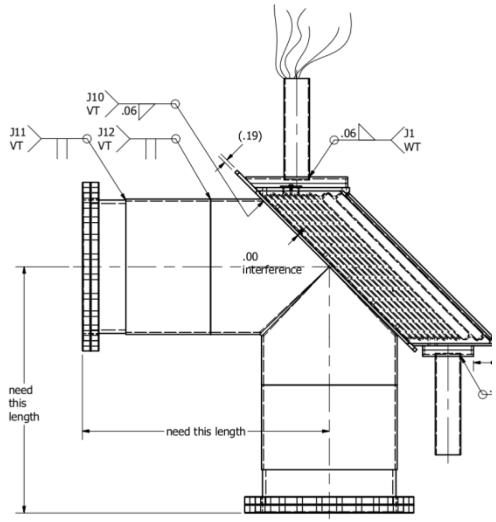
Conventional technology used for protecting accelerator vacuum systems does not preserve high quality vacuum in the accelerator systems.

Helium is a lightweight and therefore a fast-moving gas.

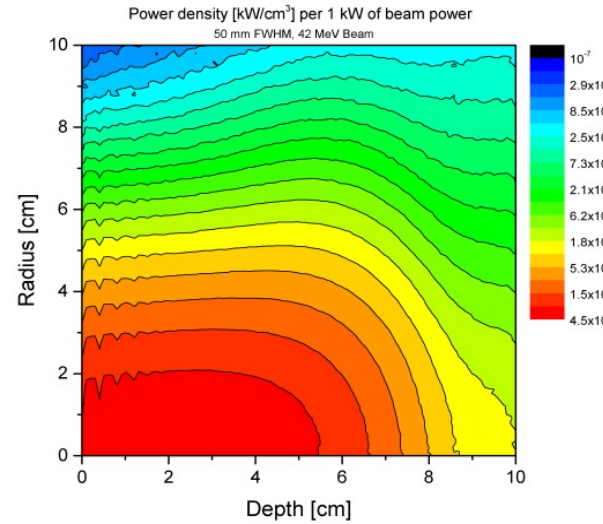
A mitigating factor for the high power system is the use of helium, as an inert gas may protect accelerator components even in the event of a pressure rise following a window failure.

HIGH POWER BEAM STOP AND COLLIMATOR

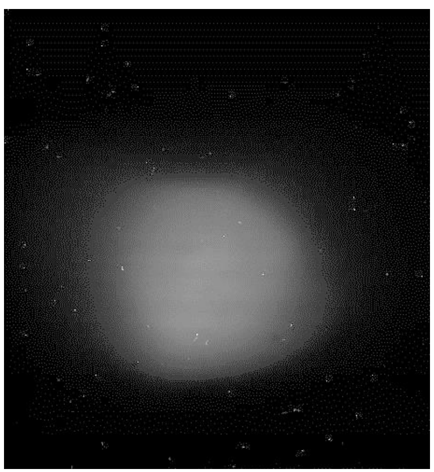




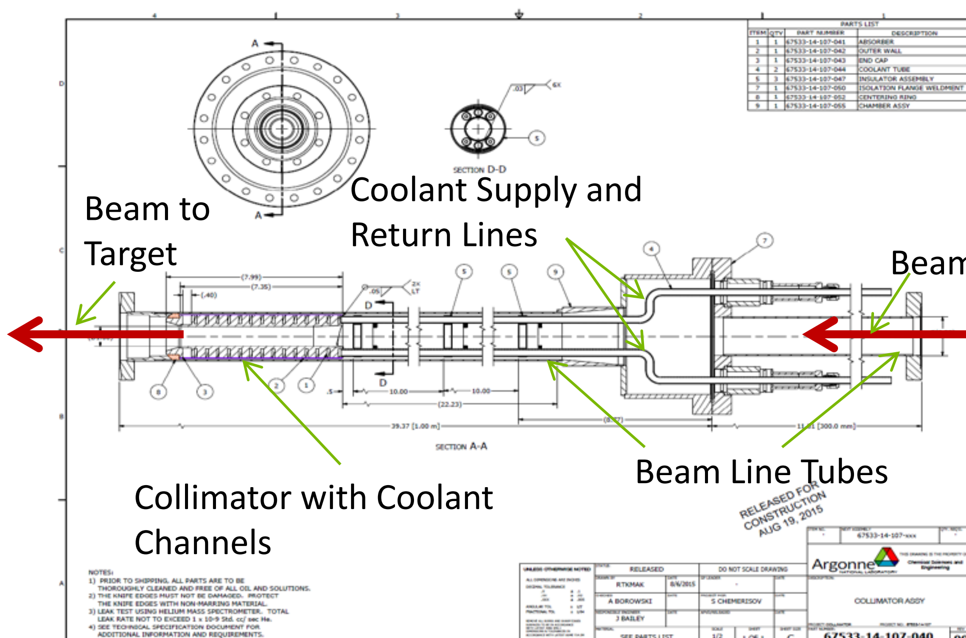
High power beam stop



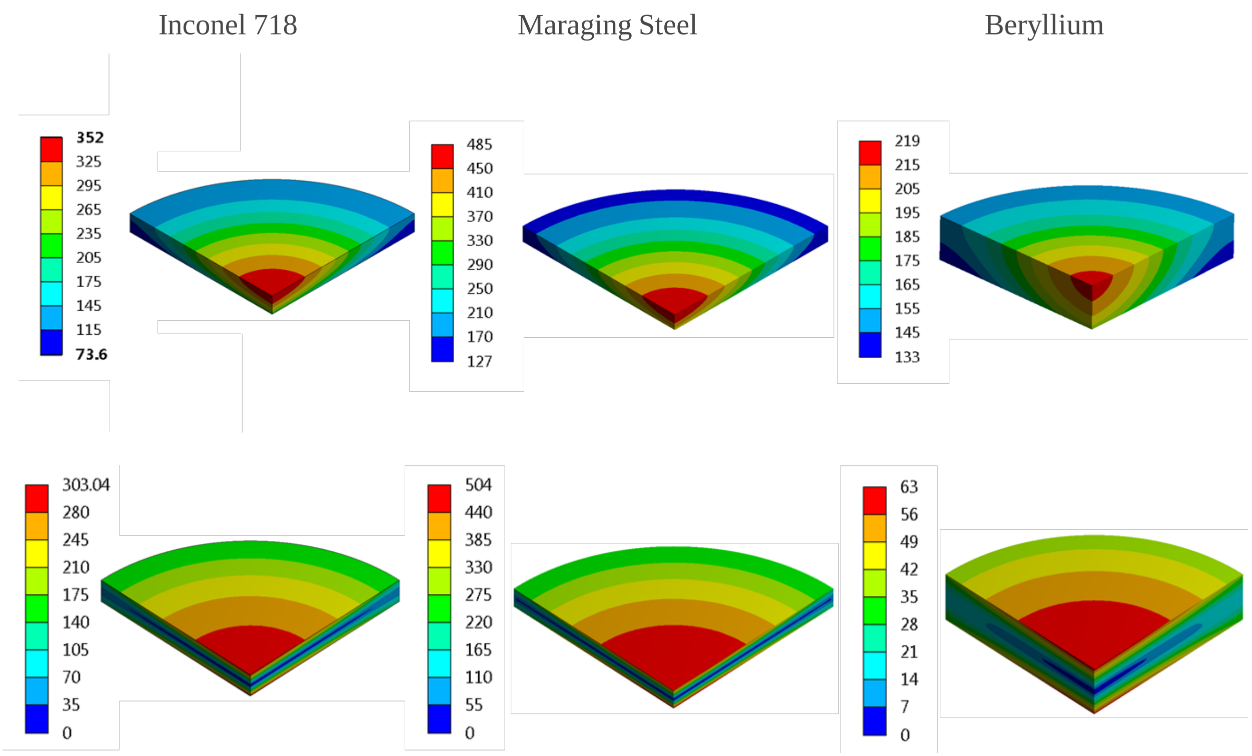
Power deposition from 42 MeV electron beam in aluminum



Defocused electron beam at the front plate of the Beam Dump



TARGET WINDOW MATERIALS CANDIDATES AND CALCULATIONS



Results of the thermal model are shown here as plots of temperature (°C)

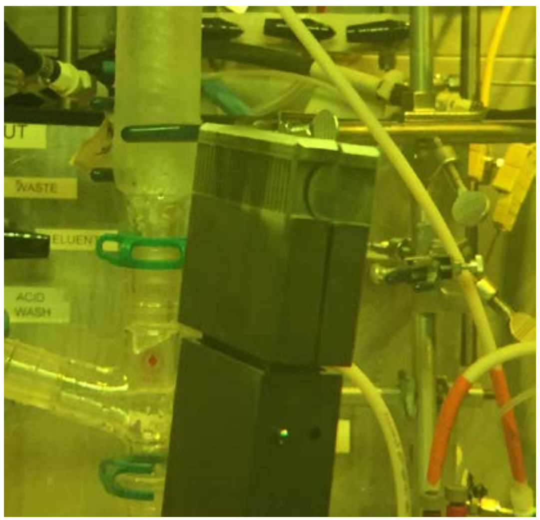
Stress due to pressure loading. Plotted as stress intensity in MPa.

Material	Maximum Beam Power (kW)
Inconel 718	18
Beryllium	40
250 Maraging Steel	45

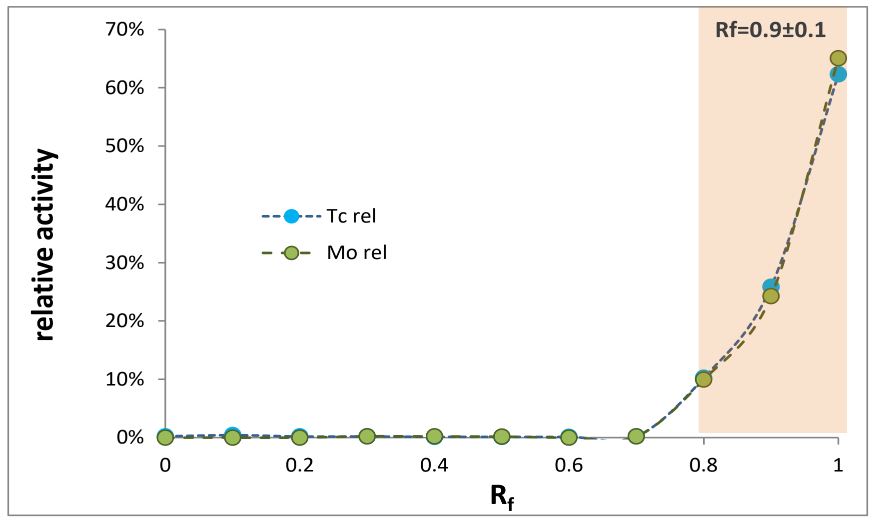
Radiation-induced damage studies are in progress

ACHIEVING RADIOCHEMICAL PURITY

In 2015-16 irradiation tests a fine orange-brown powder was observed on the irradiated Mo disks and target housing . The source of the powder was later traced to the He cooling loop. After dissolution, light orange colored solutions were obtained and radiochemical purity (RCP) tests using thin-layer chromatography (TLC) performed with the undiluted Mo solution did not meet the specifications of ≥95% of activity at $R_f = 0.9 \pm 0.1$ (R_f = retention factor).



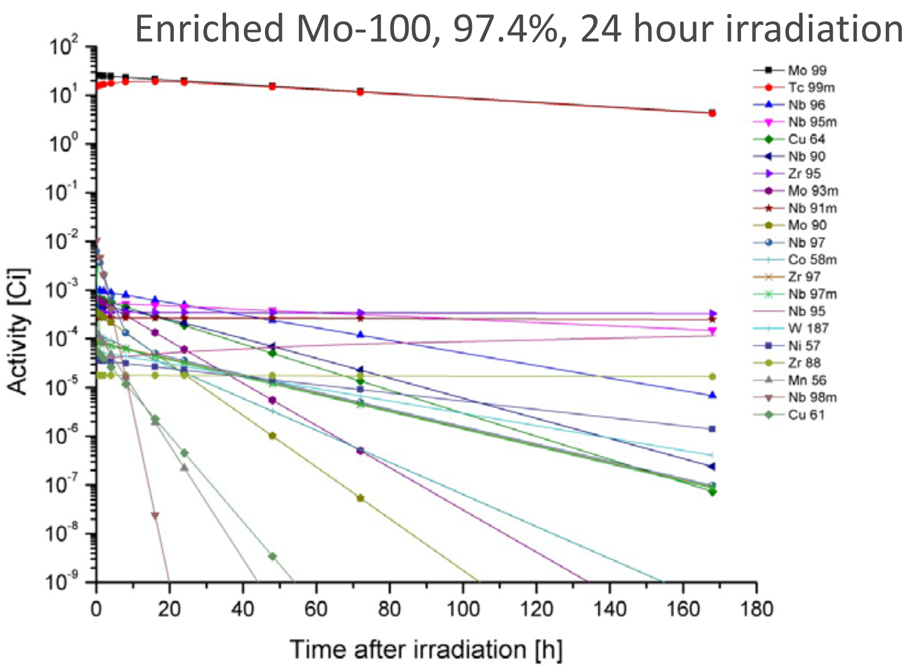
Target after irradiation

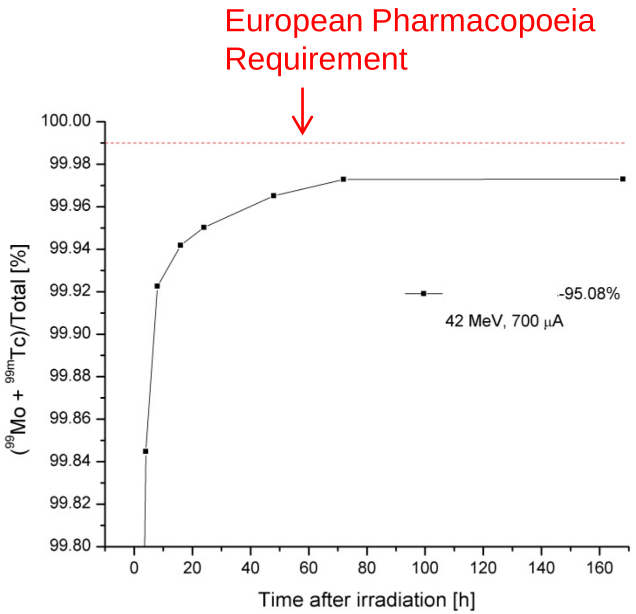


Retention factors for Mo and Tc, after spotting 20 µL of 5× diluted solution of Mo in 5M KOH on TLC and eluting in 0.1M Na_2CO_3 solution

Spotting of mobile phase (Na_2CO_3) on TLC using phenolphthalein

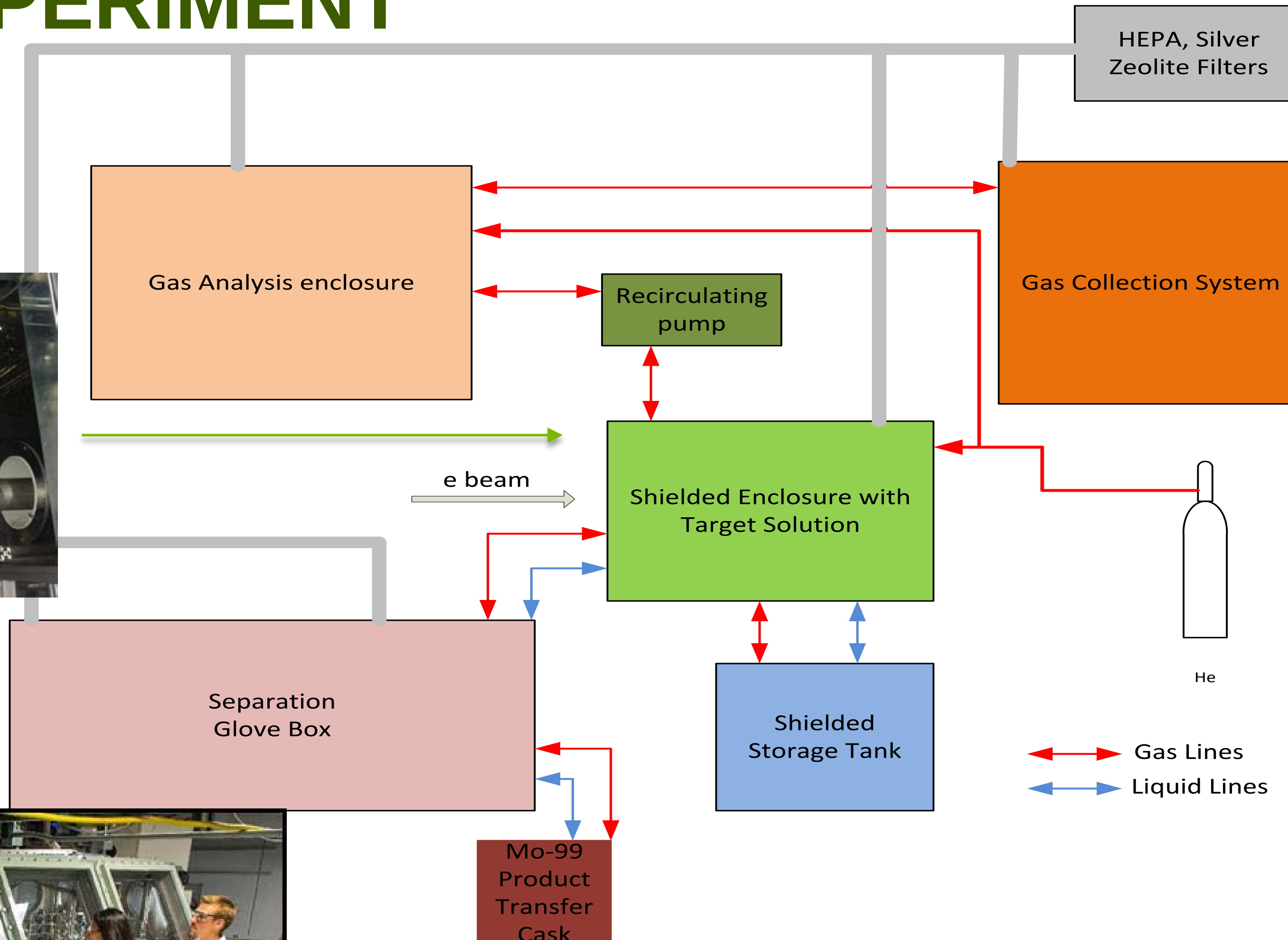
SIDE-REACTION MODELING OF ENRICHED MO-100 TARGET COMPARISON WITH EXPERIMENT





	enrichment	sample	EOB	time of counting	count duration, sec	DT
sample	enriched Mo-100	Mo100 090517	09/05/17 at 6:24PM	09/06/17 at 4:30PM	64080	20.00%
	97.40%					
Nuclide	energy, keV	T1/2, hrs	A at time of count, uCi	A at EOB, uCi	1s, %	% of Mo99 at EOB
Mo-90	257.34	5.67	2.50E-01	3.73E+00	MDA	0.0403%
Mo-93m	1477.2	6.95	3.84E-02	3.48E-01	MDA	0.0038%
Mo-99	739.5	66.19	7.34E+03	9.25E+03	3.0%	N/A
Nb-95m	235.4	86.592	3.10E-01	3.70E-01	MDA	0.0040%
Zr-95	756.7	1536.48	2.94E-01	2.97E-01	8.1%	0.0032%
Nb-95	765.8	839.52	3.25E-02	3.31E-02	MDA	0.0004%
Nb-96	1091.5	23.35	1.42E+00	2.73E+00	3.3%	0.0295%

AMORE EXPERIMENT



Experiment will be conducted at 40 MeV beam energy and up to 20 kW beam power

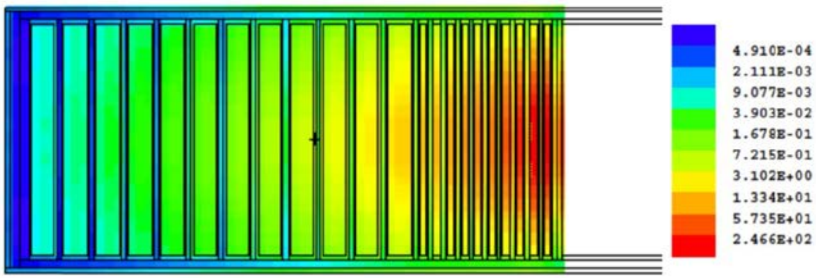
20 L solution will be irradiated with neutrons generated in a depleted-uranium (DU) target (Zr-clad DU discs were manufactured at LANL)

Maximum solution power will be ≤ 0.5 kW/L

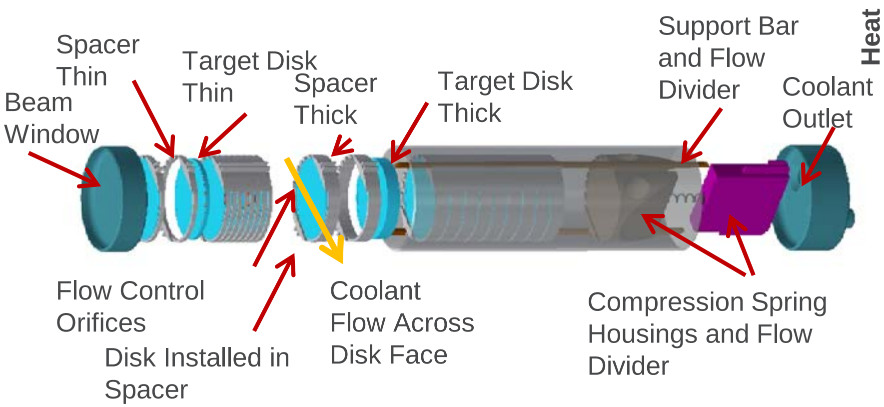
Up to 20 Ci of Mo-99 will be produced

AMORE DU TARGET DESIGN

Target

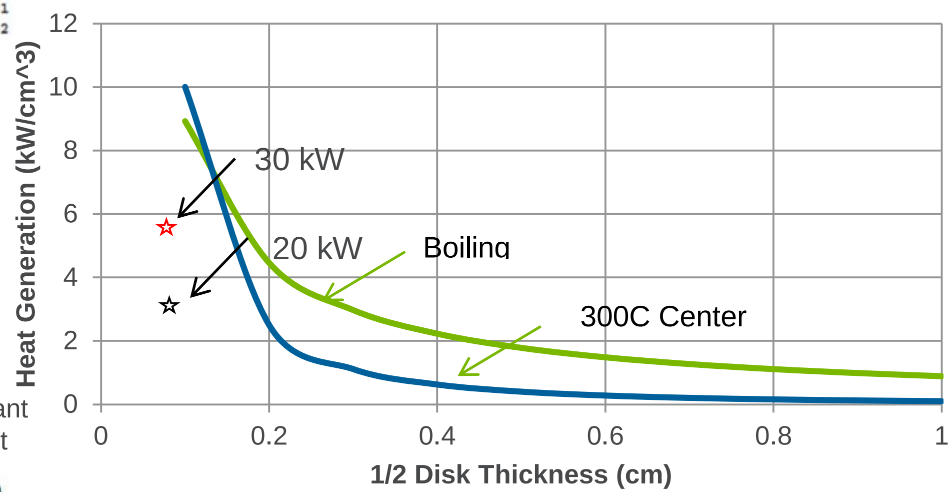


DU disks 2 mm thick
Target diameter 50 mm
Water cooling
Back disks are 6 mm thick



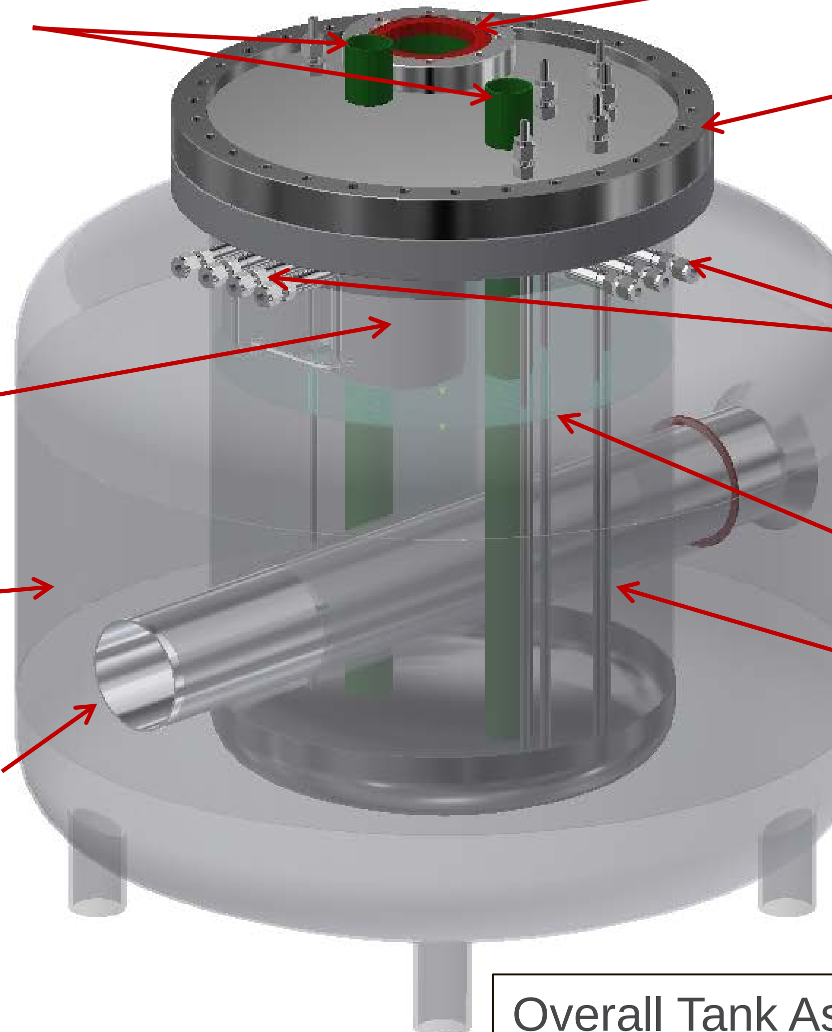
Electron beam

DU Disk Sizing



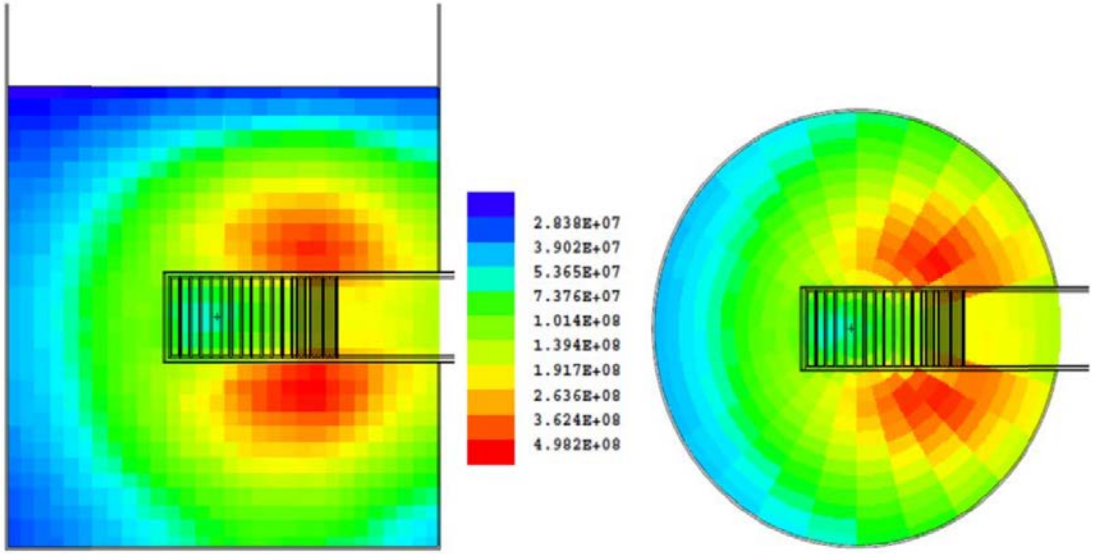
Parametric plot of heat generation vs. target half thickness for disk target geometry. The red line is defined by 300°C maximum temperature in DU disk; the blue line is defined by 100°C maximum surface temperature to prevent boiling of the coolant.

OVERVIEW OF 20L PROCESS TANK DESIGN



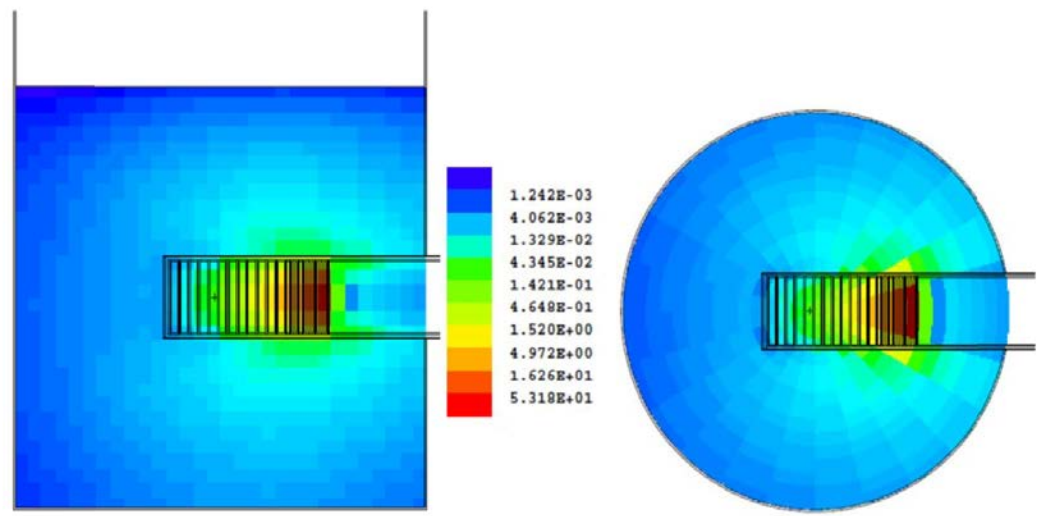
Overall Tank Assembly Size: ø22” x 22” H

PHASE 2 TARGET SOLUTION NEUTRON FLUX AND POWER DEPOSITION



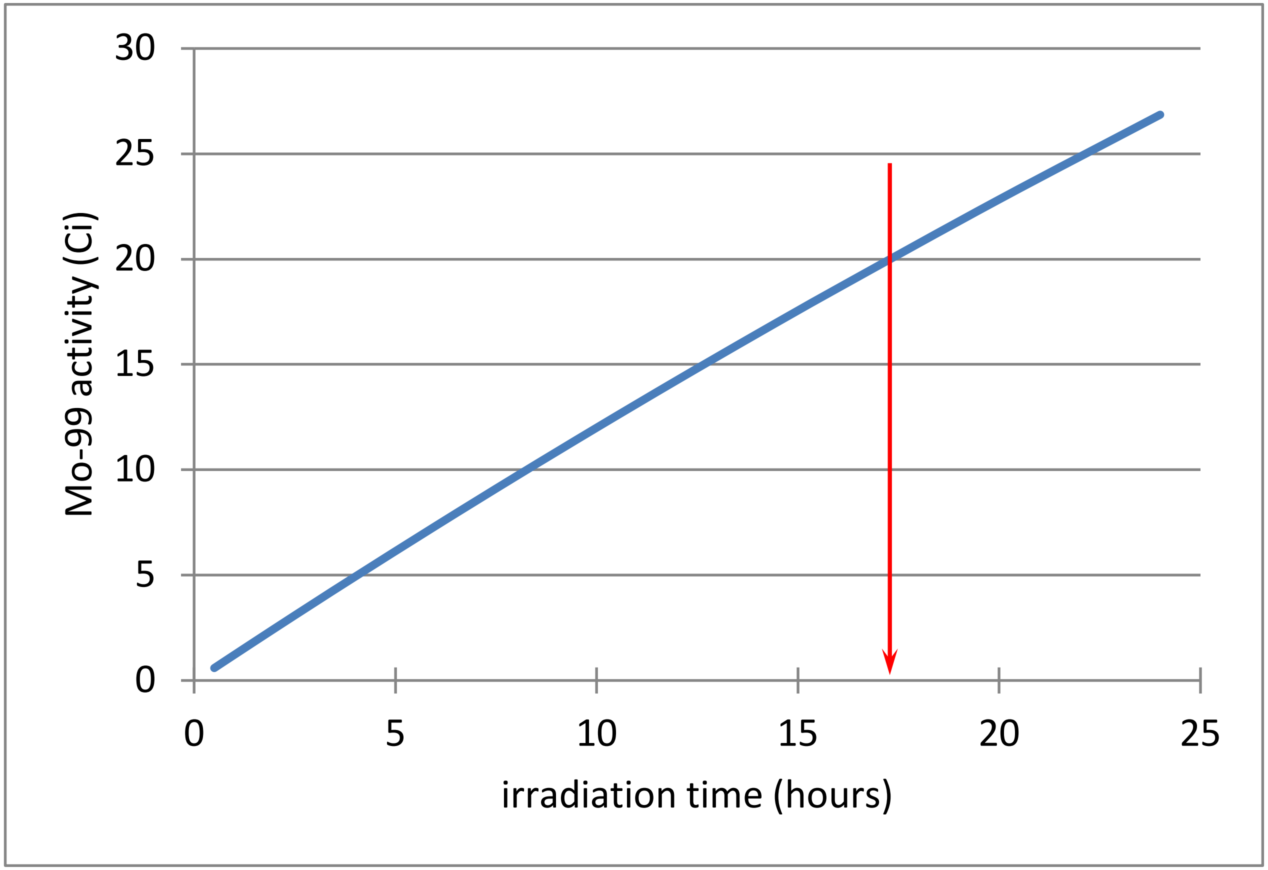
Fission rates (# fissions/cm^3/kW) in the uranyl sulfate solution, 35 MeV electron beam. Left — side view. Right – top view.

Energy deposition (watts/cm^3/kW) in the uranyl sulfate solution, for 35 MeV electron beam. Left – side view. Right – top view.



Mo-99 PRODUCTION IN AMORE EXPERIMENT

- Uranyl sulfate LEU sollutionirradiated with neutrons from the depleted uranium target
- Target production goal 20 Ci
- Estimated time to production of 20 Ci = 17.3 hours



SUMMARY

NorthStar

- Achromatic bending systems for production facility were developed and tested
- High power beam dump and collimator for production facility power was developed and tested
- New materials for target window were proposed. Testing of the materials is underway
- Source of contamination in helium cooling system was identified and eliminated. Mo-99 produced after clean-up met radiochemical purity specification
- Experimental verification of side reactions showed better then expected Mo-99 purity

AMORE experiment

- Complete experimental setup for irradiation of 20 L of uranyl sulfate solution is assembled.
- We are in process of commissioning of the experiment

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- Brad Micklich
- Lohman Hafenrichter
- Kurt Alford
- Ken Wesolowski
- Kevin Quigley
- Jim Bailey
- Tom Brossard
- David Rotsch

- This work is conducted in collaboration with LANL and ORNL

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