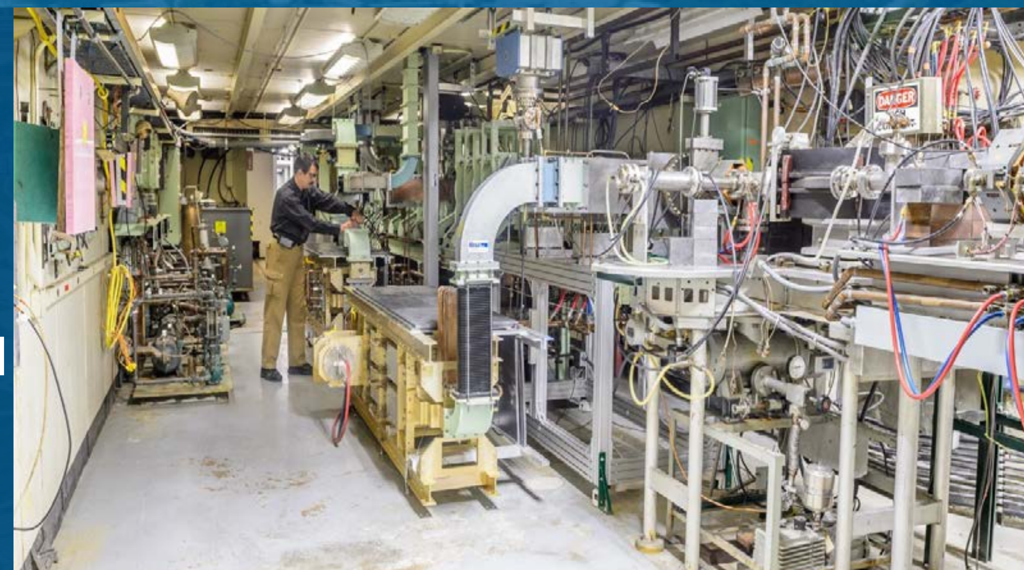
MO-99 TOPICAL MEETING

ACCELERATOR-BASED PRODUCTION OF MO-99: PHOTONUCLEAR APPROACH



SERGEY CHEMERISOV

Experimental Operations and Facilities Division

PETER TKAC, ROMAN GROMOV, JERRY NOLEN, JEONGSEOG SONG, CHARLES JONAH, AND GEORGE VANDEGRIFT

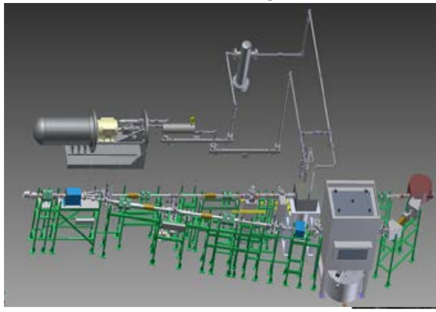
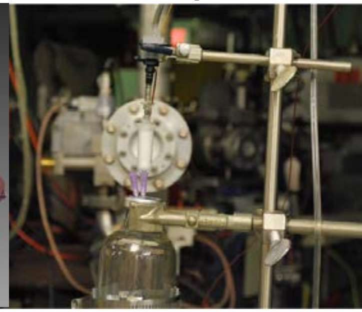
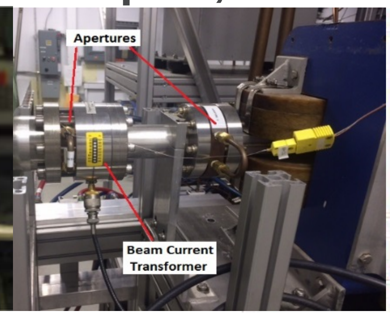
September 26, 2018
Knoxville, TN
Mo-99 Topical meeting

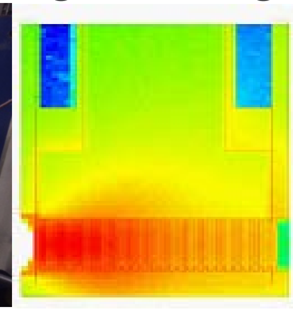
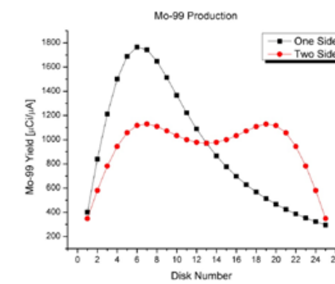
OUTLINE

- Introduction
- Target design and bremsstrahlung converter
- Window material selection considerations
- Side reaction study for enriched Mo-100
- Facility beamline and vault design

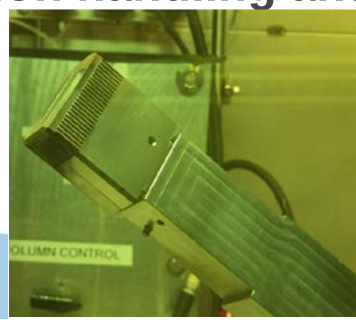
ARGONNE'S DEVELOPMENT OF ACCELERATOR-BASED PRODUCTION OF MO-99

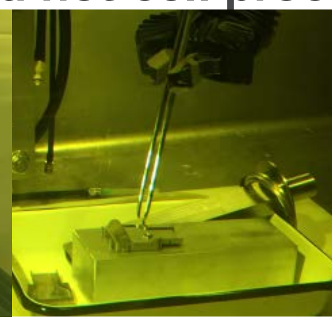
Irradiations, radiation dose, beam transport, shielding and target design, MCNPX

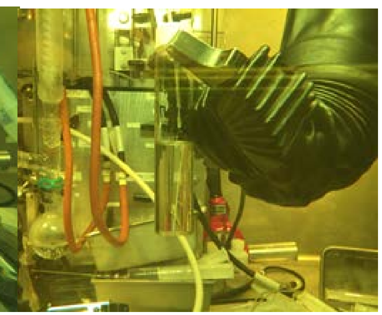




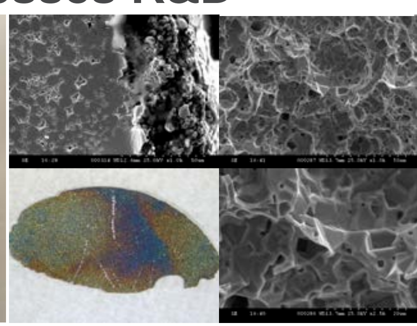
Post-irradiation handling and hot-cell processing





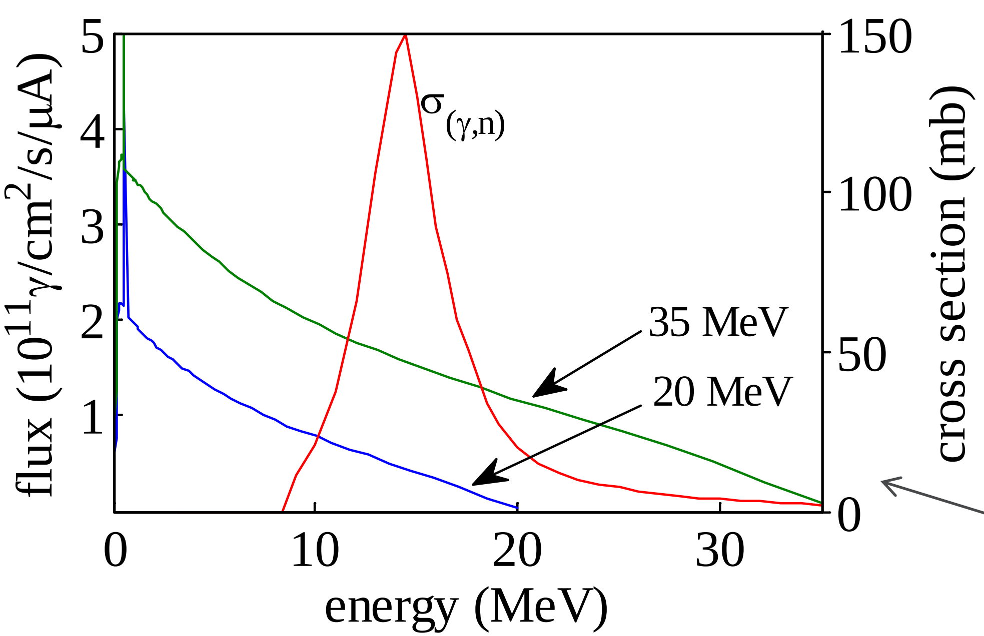
Chemical processes R&D

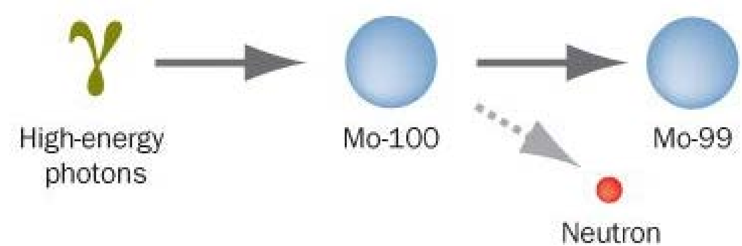





PROOF OF CONCEPT DEMONSTRATIONS FOR ELECTRON ACCELERATOR PRODUCTION OF ^{99}MO

- Under the direction of the NNSA, ANL and LANL are partnering with NorthStar Medical Radioisotpes. to demonstrate and develop 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.
- Enriched ^{100}Mo should be commercially available for $500–1000 per gram for kg quantities.

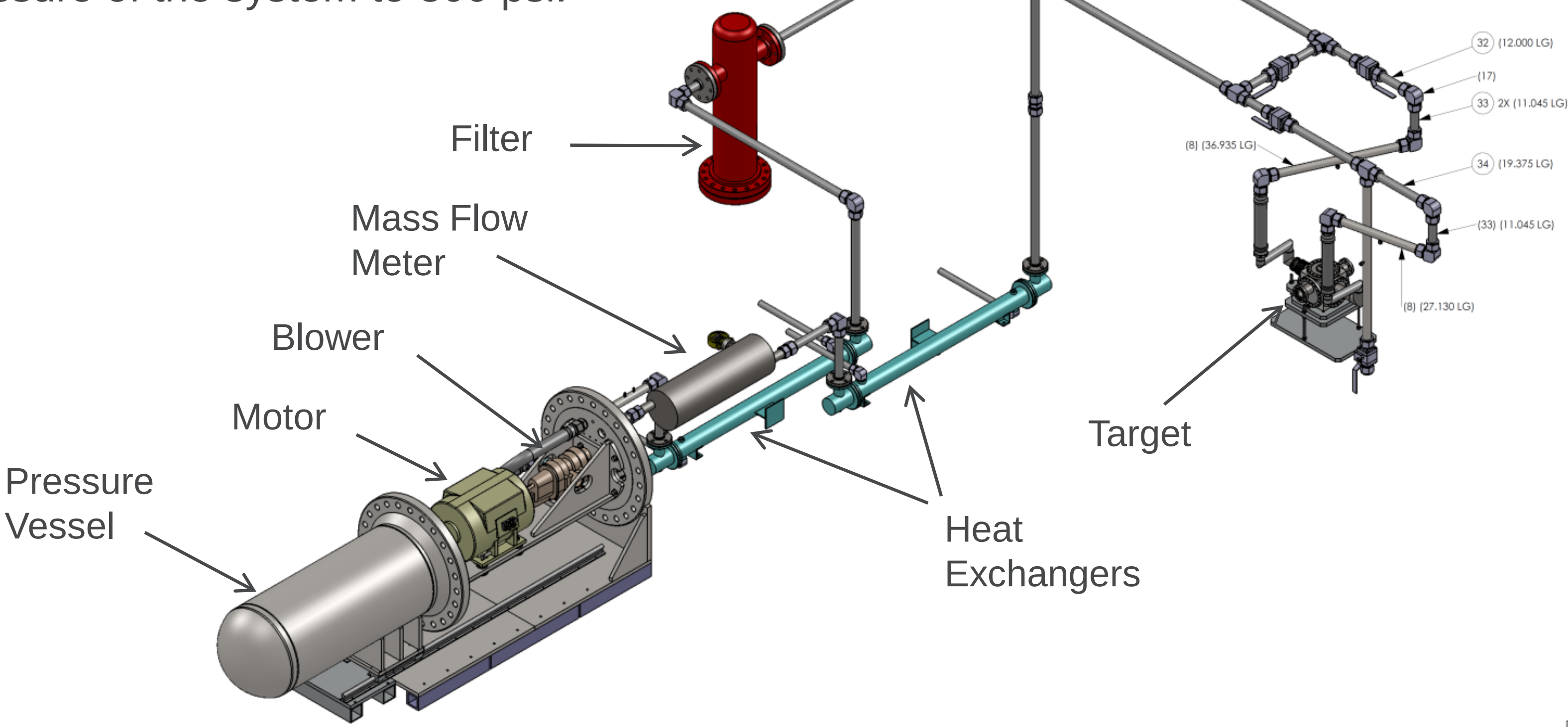




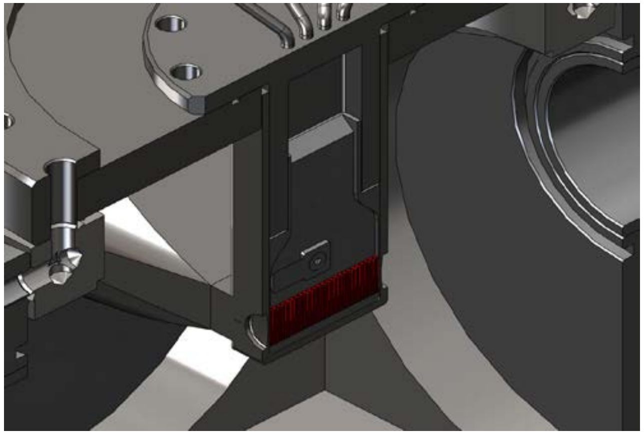
Comparison of the bremsstrahlung photon spectra produced with 20- and 35-MeV electron beams in a Mo target compared with photonuclear cross section of ^{100}Mo.

CLOSED LOOP GASEOUS HELIUM COOLING SYSTEM LAYOUT AT ARGONNE

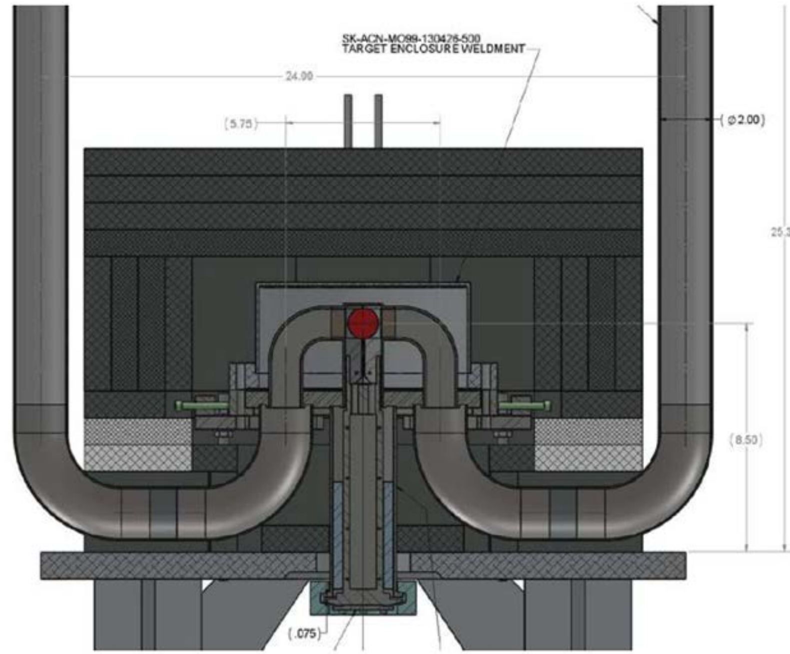
The roots blower is used to move the He through the loop and across the targets. The PV is used to increase the base pressure of the system to 300 psi.



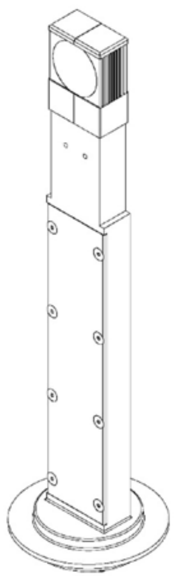
TARGET DESIGN

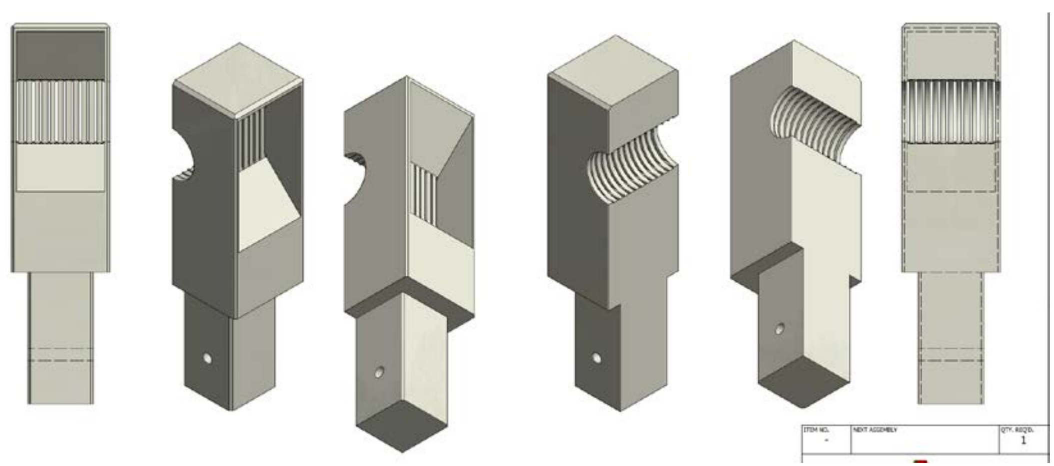


First 12 mm target



29 mm target

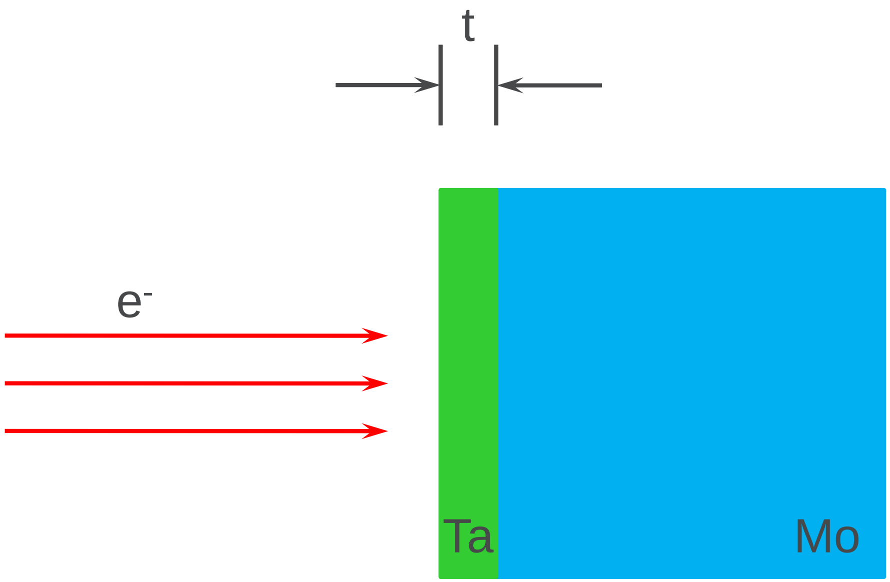




29 mm insert to hold 12 mm disks

CONVERTER STUDY FOR ^{99}MO TARGET

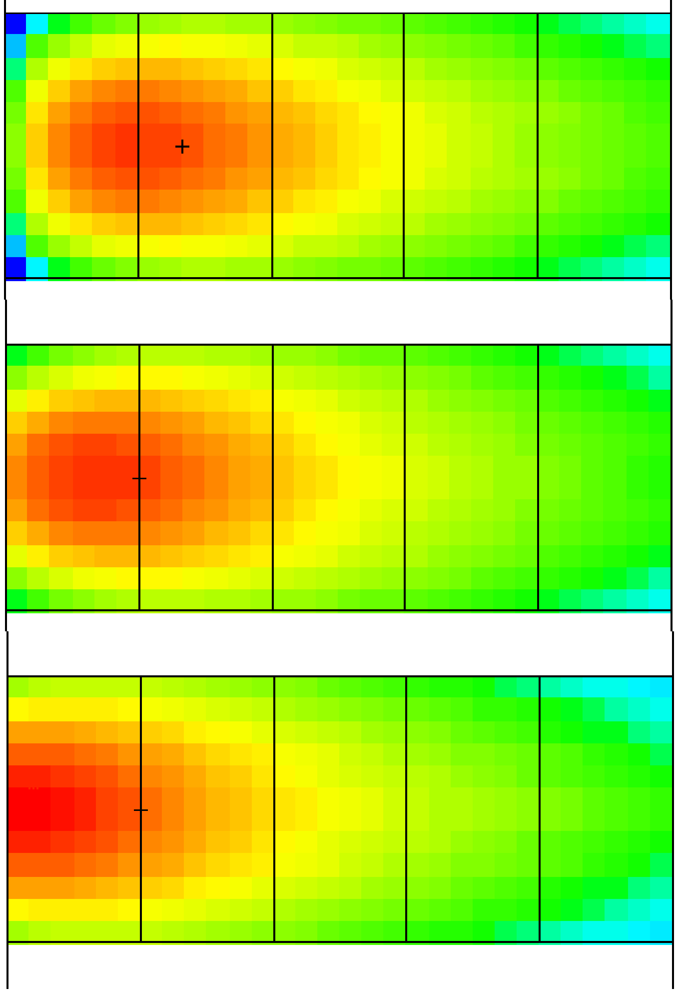
- Would use of the high-Z (e.g., Ta) converter in front of the moly target increase the ^{99}Mo yield by increasing the conversion of electron energy to photons?



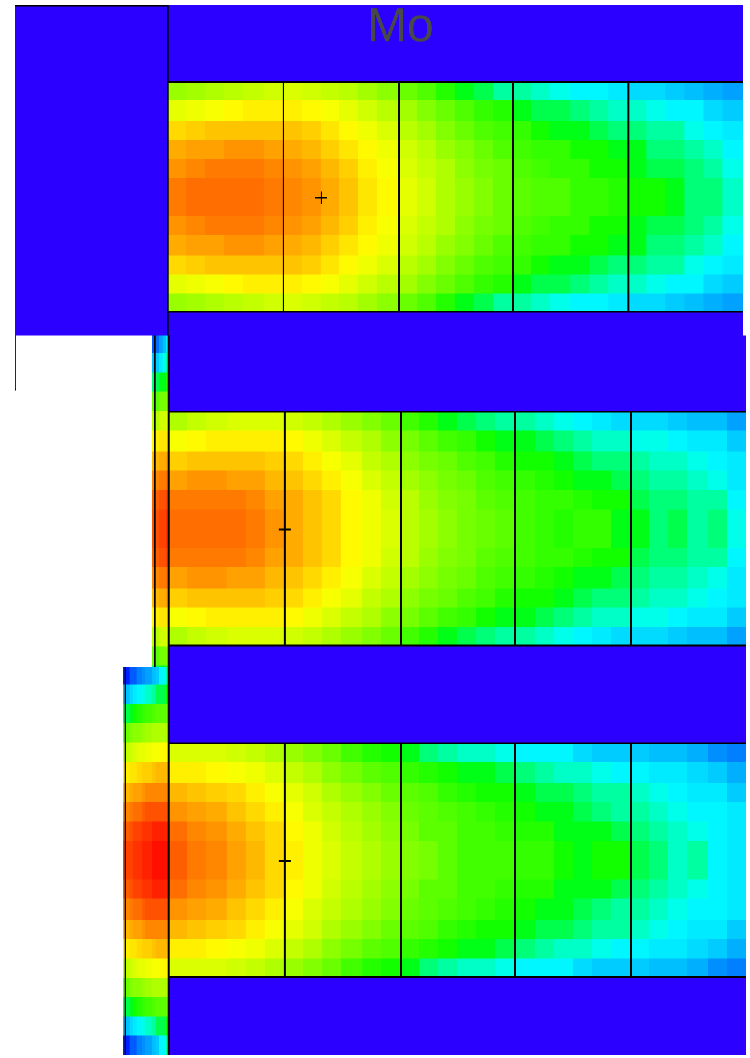
CONVERTER STUDY FOR ^{99}MO TARGET

- Electron beam (35 MeV) incident from the left

^{100}Mo(n,)^{99}Mo rate



Energy deposition in Mo



no Ta
Mo yield
1.0

0.077 cm Ta
Mo yield
1.06

0.231 cm Ta
Mo yield
1.04

MATERIALS SELECTION FOR HIGH POWER TARGET

Material	Density (Kg/m³)	Thermal Conductivity (W/m-°C)	Maximum Stress (MPa)	Minimum Window Thickness (mm)	Maximum Temperature (°C)	Figure of Merit (FOM)
INCONEL 718	8,221	17.3	456	1.15	403	1
Hastelloy X	8,221	26.0				*Disqualified
INCONEL 706	8,055	22.5	75	2.87	1,280	2.45
Waspaloy	8,193	17.3	357	1.30	481	1.13
Rene 41	8,249	17.3	507	1.09	388	0.96
L-605 Haynes Alloy 25	9,134	19.0				*Disqualified
316 SS	7,806	22.5				*Disqualified
250 Maraging Steel	7,916	29.4	706	0.93	269	0.78
AerMet 100	7,889	31.2	793	0.87	249	0.73
2024-T81 Aluminum	2,768	173.1				*Disqualified
6061-T6 Aluminum	2,713	173.1				*Disqualified
Titanium alloy AMS 4910	4,484	13.9	175	1.88	497	0.90
Beryllium Standard grade	1,855	138.5	147	1.96	131	0.39
Magnesium alloy	1,800	77.0				*Disqualified

$$FOM = \frac{\rho t}{\rho_I t_I}$$

ρ = density of material to be evaluated

t = minimum acceptable thickness of material to be evaluated

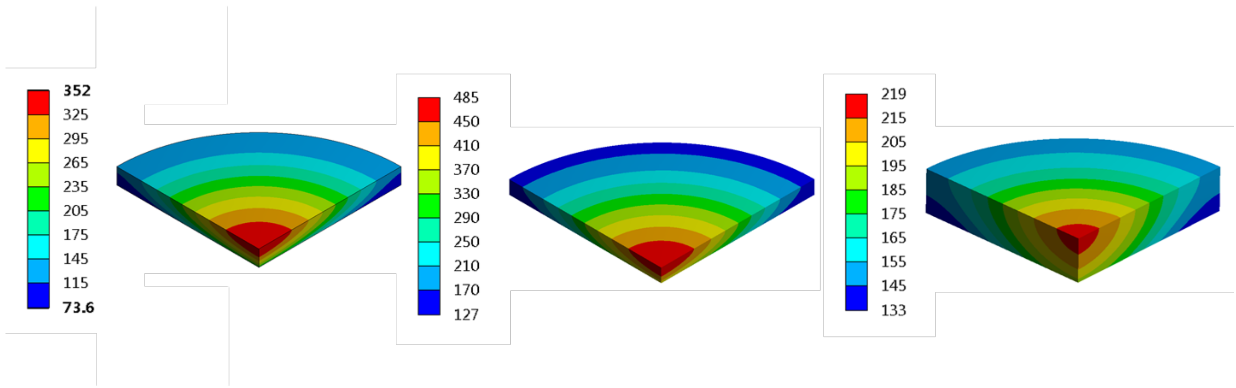
ρ_I = density of INCONEL 718

t_I = minimum acceptable thickness of INCONEL 718

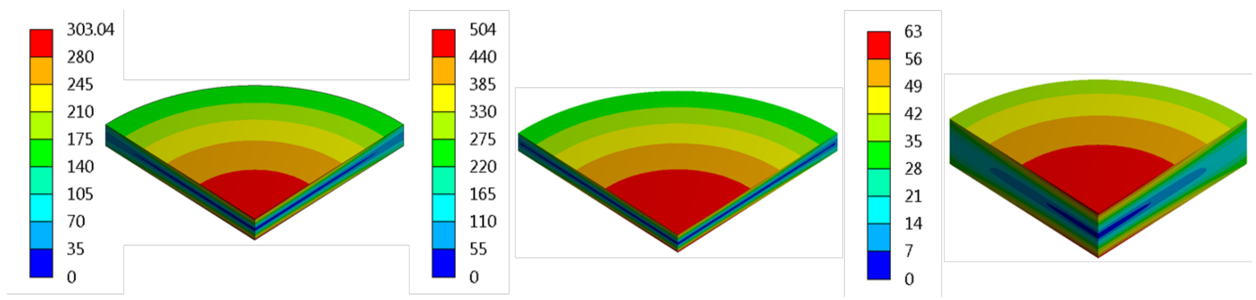
FOM = Factor of Merit

FINAL CANDIDATES AND CALCULATIONS

Inconel 718 | Maraging Steel | Beryllium



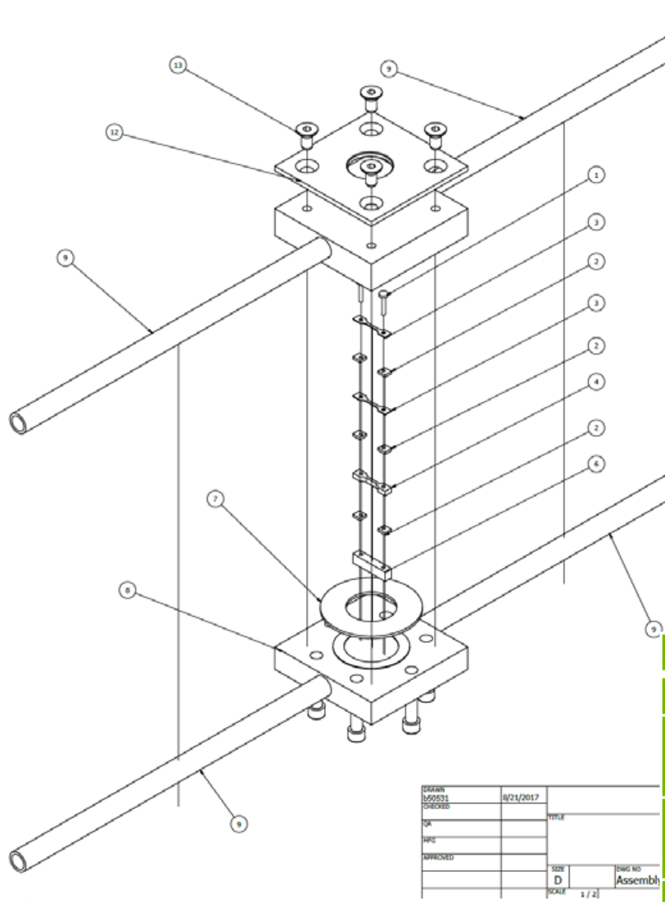
Results of the thermal model are shown here as plots of temperature (°C)

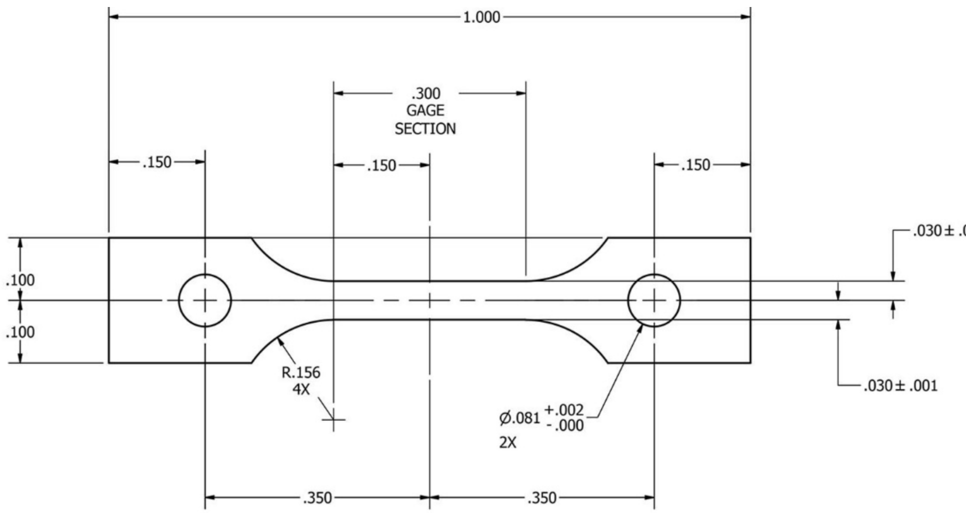


Stress due to pressure loading. Plotted as stress intensity in MPa.

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

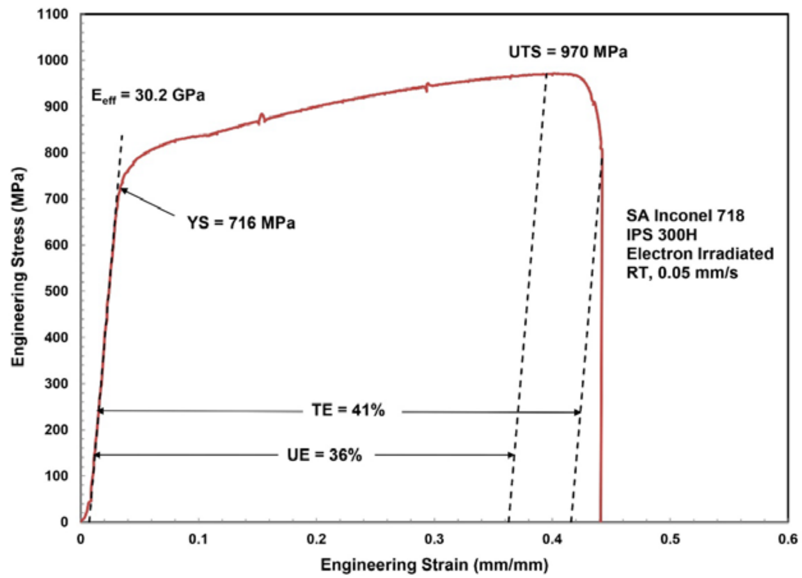
TESTING WINDOW MATERIALS CANDIDATES

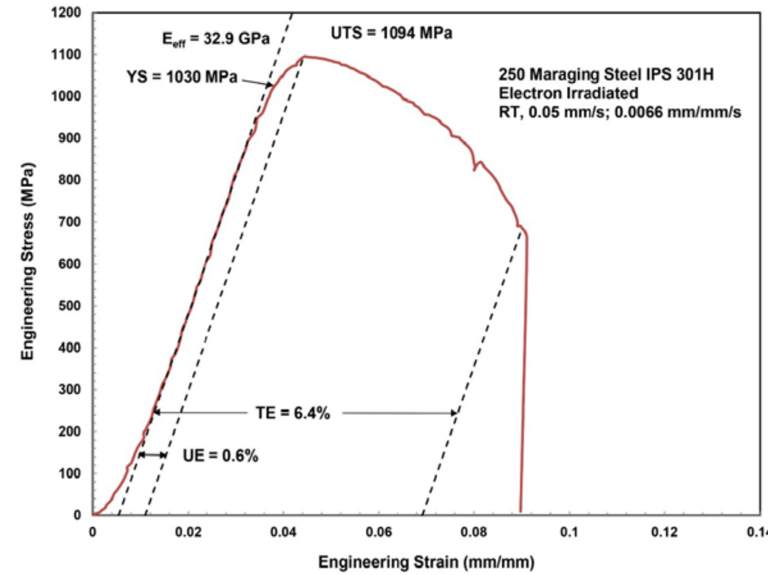


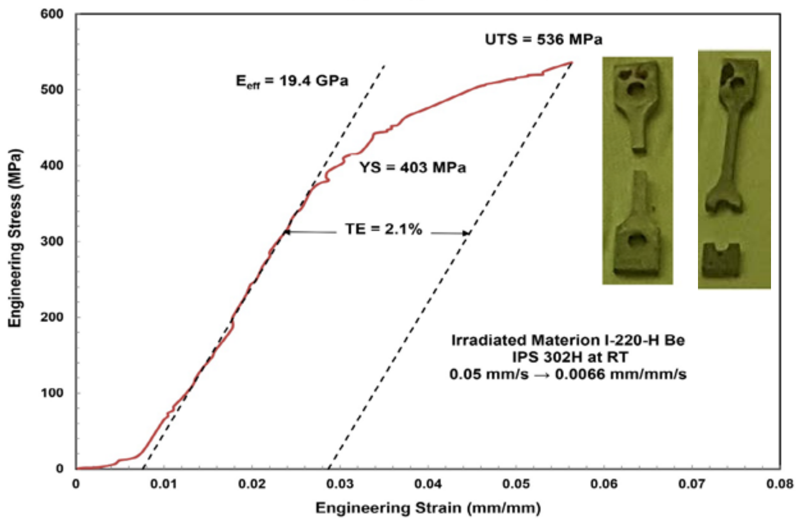


Parameter	IN 718	MS	Be
Gauge Length, in. (mm)	0.300 (7.62)	0.300 (7.62)	0.300 (7.62)
Gauge Width, in. (mm)	0.060 (1.542)	0.060 (1.542)	0.060 (1.542)
Gauge Thickness, in. (mm)	0.020 (0.508)	0.020 (0.508)	0.060 (1.542)
Total Length, in. (mm)	1.000 (25.40)	1.000 (25.40)	1.000 (25.40)
Yield Stress, ksi (MPa)	61.5 (424)	252 (1738)	50 (345)
Ultimate Tensile Stress, ksi (MPa)	130.5 (900)	257 (1772)	65 (448)
Uniform Elongation, %	---	---	---
Total Elongation, %	51.8	9.0	2.0
Reduction in Area, %	---	63	---

TENSILE TESTING RESULTS





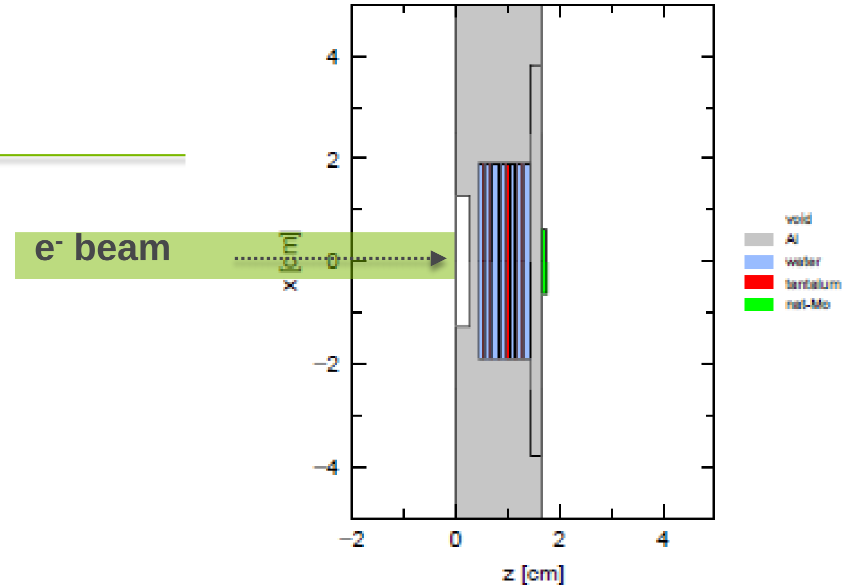


Material	Baseline YS, Mpa	Baseline UTS, Mpa	Baseline TE, %	Irradiated YS, Mpa	Irradiated UTS, Mpa	Irradiated TE, %
Inconel 718	488	901	64	662	957	38
Maraging Steel 250	882	990	9	1021	1076	6
Beryllium	489	546	2.9	403	536	2.1

YS – Yield Stress, TE – Total Elongation

UTS – Ultimate Tensile Stress

EXPERIMENTAL SETUP FOR SIDE-REACTIONS STUDY



Beam:

40 MeV, 1.5 kW power

30 min, 4 hrs

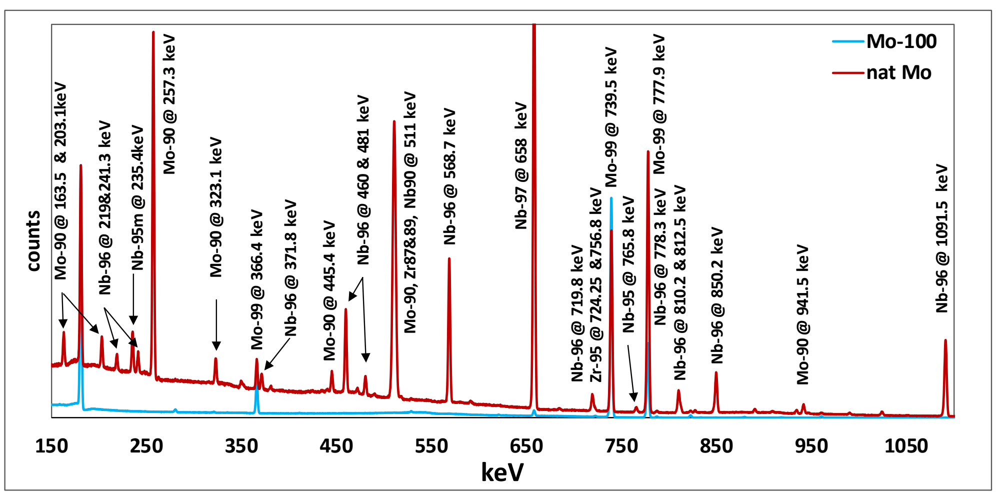
Target:

Nat and enriched ^{100}Mo-97.4%, 2.6% ^{98}Mo)

Ta convertor 3 mm (6×0.5 mm) water cooled

Al plates before and after Convertor: ~3mm thick

2 Mo targets 1 mm thick each



Enriched Mo100

Natural Mo

Impurities

Nuclide	energy, keV	T1/2, hrs
^{90}Mo	257.3	5.67
^{99}Mo	739.5	66.2
^{90}Nb	1129.1	14.6
^{91m}Nb	1205	1536.1
^{92m}Nb	934.5	243.8
^{95m}Nb	235.4	86.6
^{95}Nb	765.8	839.5
^{96}Nb	1091.5	23.4
^{97}Nb	657.9	1.23
^{98m}Nb	787.2	0.852
^{88}Zr	392.85	2001.6
^{89}Zr	909.2	78.4
^{95}Zr	724.18	1536.5
^{88}Y	1836&898	2558.4
^{51}Cr	320.07	664.8
^{54}Mn	834.8	7490.4
^{57}Co	122.1	6480

SIDE-REACTION PRODUCTS ON ENRICHED TARGET (97.4% Mo-100, 2.6% Mo-98)

Short lived:

^{98}Mo(γ,pn)^{96}Nb - 23.35 hrs
^{98}Mo(γ,p)^{97}Nb – 1.23 hrs
^{100}Mo(γ,pn)^{98m}Nb – 0.852 hrs

Long lived:

^{98}Mo(γ,p2n)^{95}Nb – 840 hrs
^{100}Mo(γ,n)^{99}Mo – 66.2 hrs
^{100}Mo(γ, αn)^{95}Zr – 1536 hrs

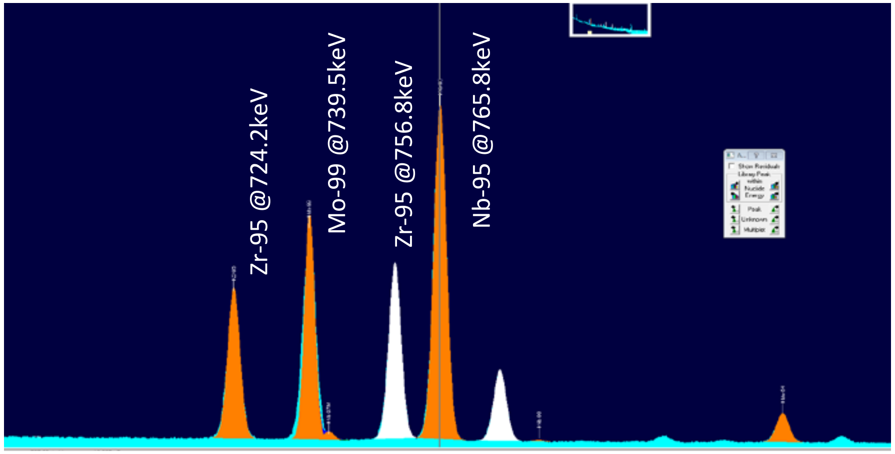
97.4% Mo100 2.6% Mo98	ppm
W	75.1
Ge	11.4
Cu	14.9
Ni	39.4
Fe	540
Mn	5.7
Cr	64

^{185}W – lbr, ^{181}W – <70keV, ND
^{71}Ge – 10keV, ^{69}Ge – 511keV
^{64}Cu – 511keV

^{55}Fe – 6keV

Impurities:

^{55}Mn(γ,n)^{54}Mn – 7490 hrs
^{52}Cr(γ,n)^{51}Cr – 665 hrs
^{58}Ni(γ,p)^{57}Co – 6480 hrs

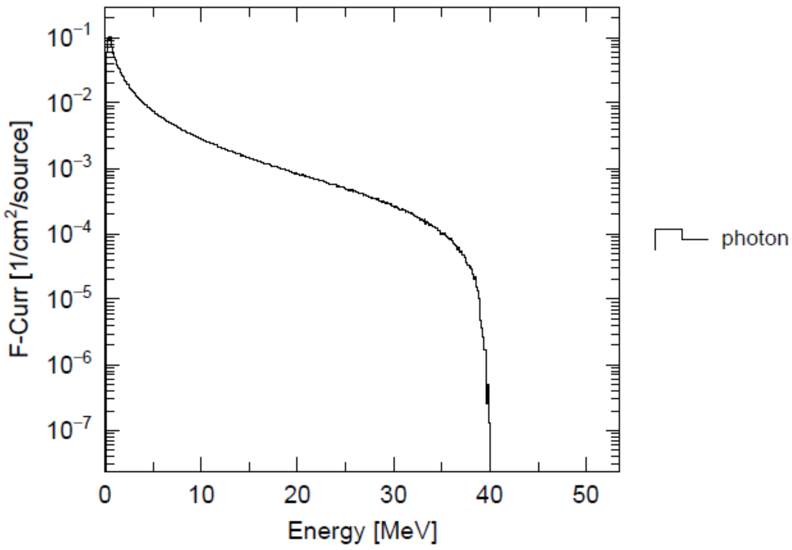


No ^{95m}Nb detected – low production

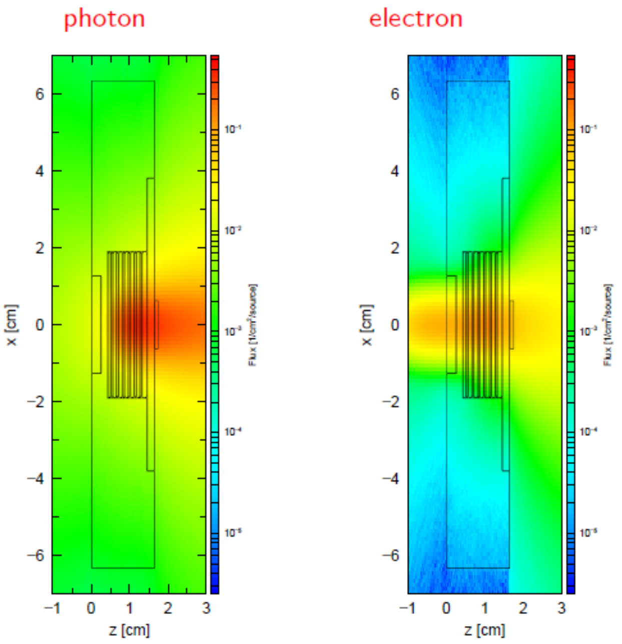
MONTE CARLO CALCULATIONS

Monte Carlo simulation tool: PHITS 3.02
Photonuclear reaction cross sections: JENDL

- γ ray energy distribution on the target
  - ✓ Energy vs Flux



- electron and photon tracks

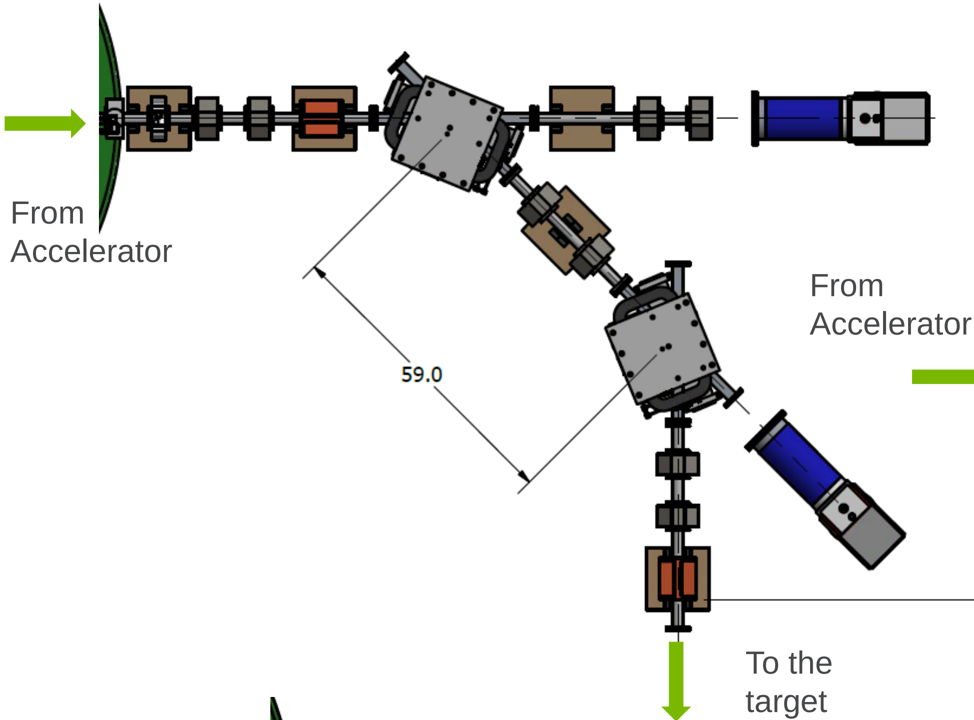


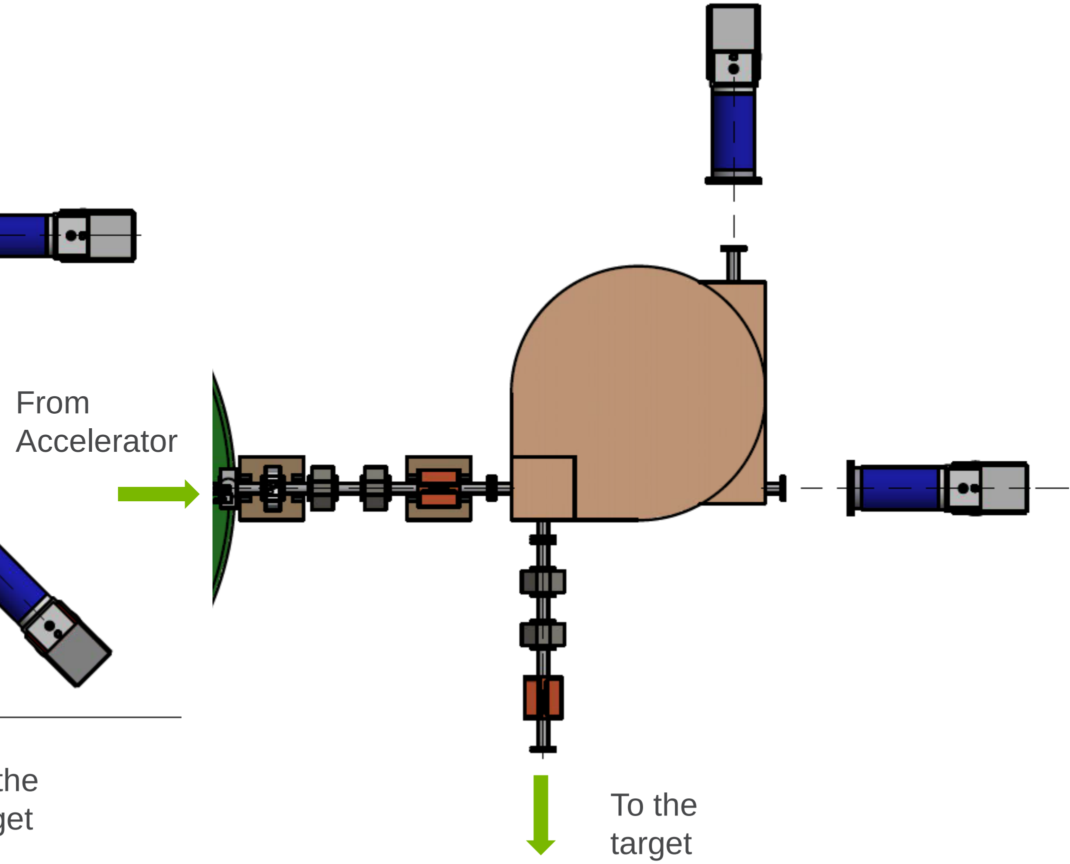
COMPARISON OF THE PRODUCTION RATES

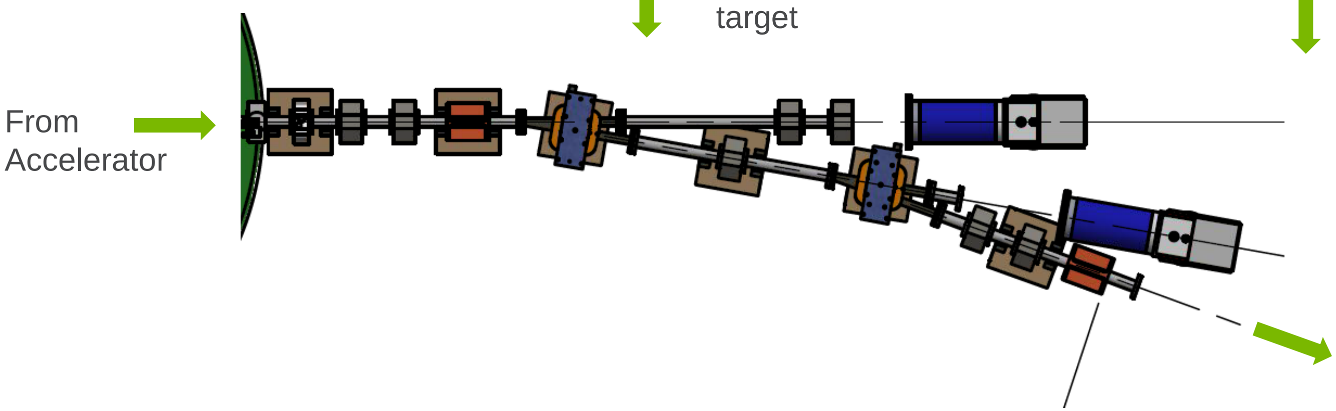
Experimental vs calculated values for 30 min and 4 hrs irradiation with enriched ^{100}Mo (97.4%)

Halflife, hours	Isotope	Experimental production rates for 30 min irradiation normalized by Mo-99 production rate	Experimental production rates for 4 h irradiation normalized by Mo-99 production rate	Calculated production rates for 30 min irradiation normalized by Mo-99 production rate	Calculated production rates for 4 h irradiation normalized by Mo-99 production rate
66.19	Mo-99	1.00E+00	1.00E+00	1.00E+00	1.00E+00
839.52	Nb-95		4.26E-05	1.42E-06	1.50E-06
23.35	Nb-96	1.12E-04	1.07E-04	1.26E-05	1.26E-05
1.233	Nb-97	1.46E-03		1.12E-04	1.13E-04
0.852	Nb-98	1.11E-03		1.56E-04	1.56E-04
1536.48	Zr-95	2.07E-04	2.02E-04	7.11E-05	7.12E-05
664.8	Cr-51		1.37E-04		
7490.4	Mn-54	3.16E-05	3.09E-05		
6480	Co-57	4.31E-05	6.00E-05		

BEAMLINE DESIGN







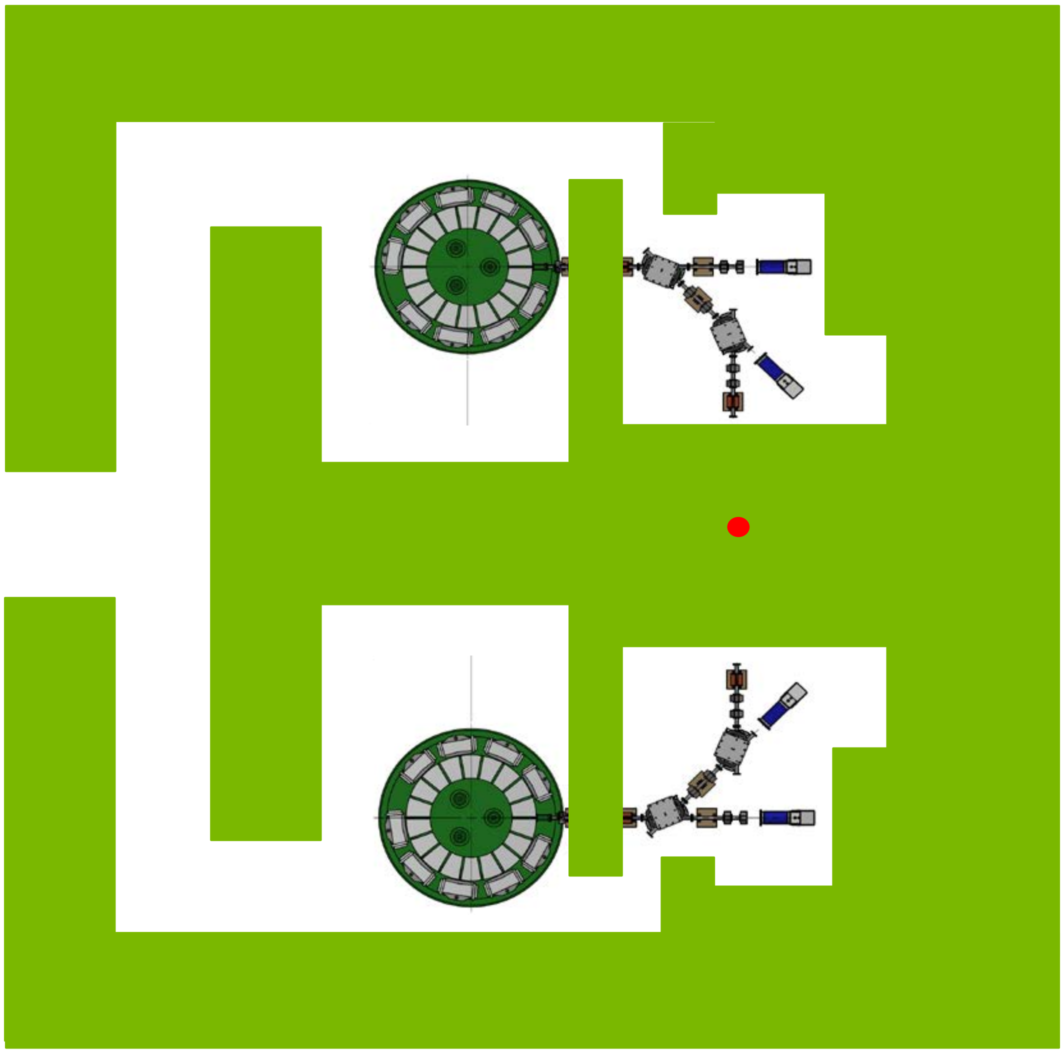
ACCELERATOR VAULT DESIGN

Requirement:

- Be able to perform maintenance on one of the accelerators while other is performing irradiation

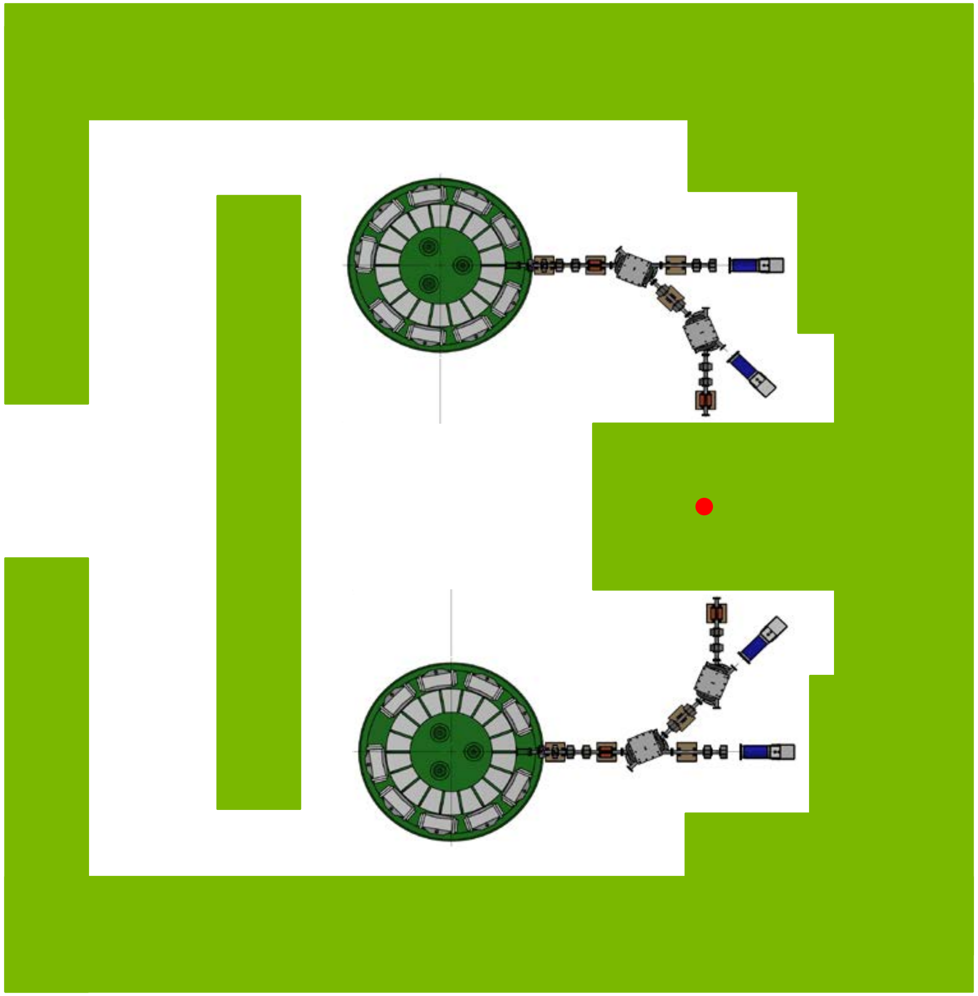
- Concrete thickness in direction of beam has to be ~4m if only ordinary concrete is used. It can be significantly reduced if lead, iron or heavy concrete is used

- 2.5 m of ordinary concrete is required on direction perpendicular to the beam

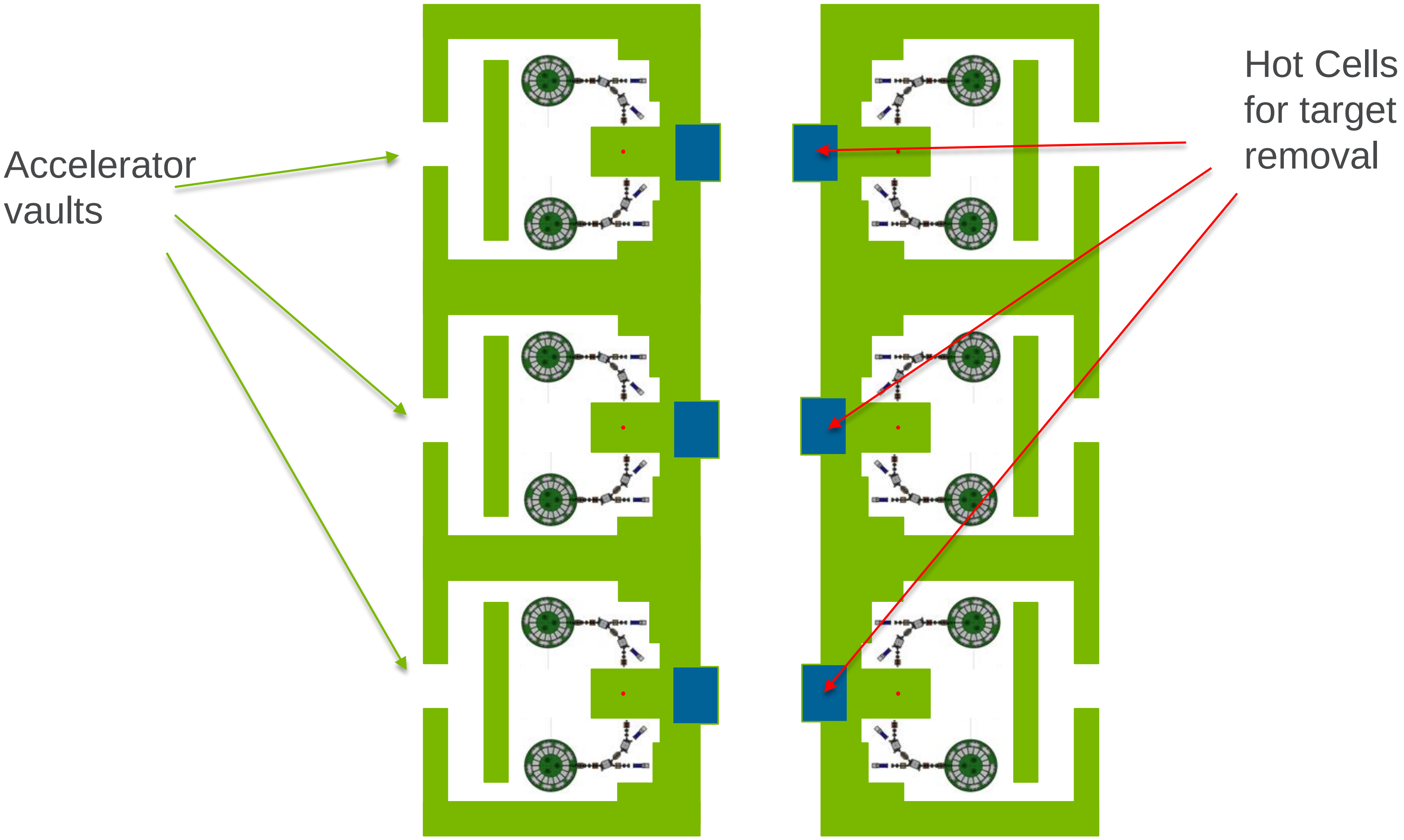


ACCELERATOR VAULT DESIGN

- When maintenance is not performed during irradiation vault can be much smaller
- Better access to the beamline and accelerator
- Shorter beamline can be used



FACILITY DESIGN



SUMMARY

- Utilization of high-Z converter provides up to 6% boost in Mo-99 production
- Beryllium and maraging steel target window can accommodate high beam power for the same target design compared with Inconel 718
- Main long-lived RN on enriched target: ^{95}Zr, ^{95}Nb
- Level of impurities introduced during recycling is important for final material purity
- Recommendations for the beamline and shielding configuration are developed

ACKNOWLEDGEMENTS

- Peter Tkac
- Roman Gromov
- Chuck Jonah
- Brad Micklich
- Kurt Alford
- Ken Wesolowski
- Kevin Quigley
- Jim Bailey
- George Vandergrift

- This work is conducted 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.