Accelerator Based Domestic Production of ^{99}Mo: Photonuclear approach

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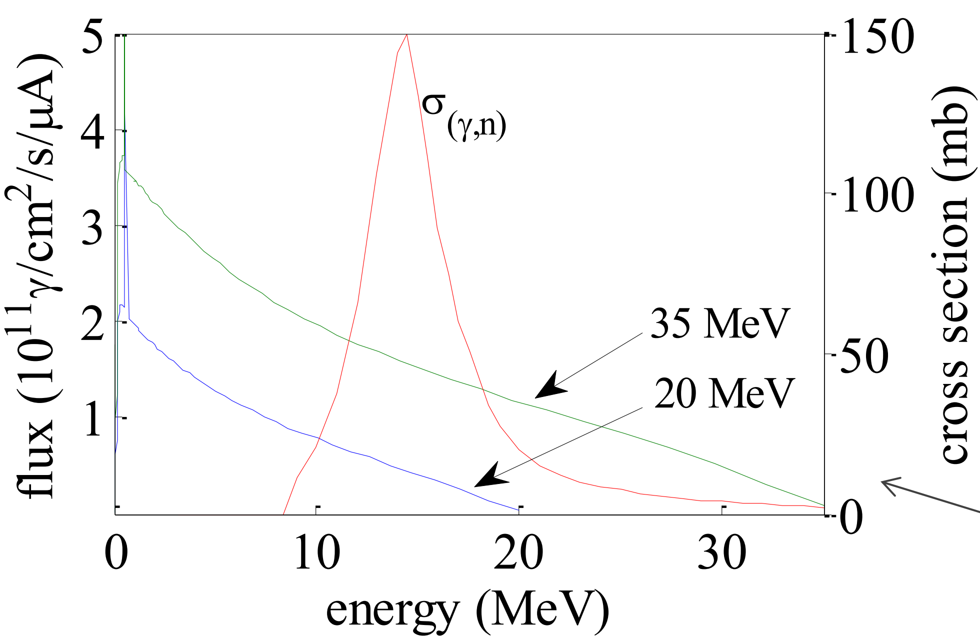
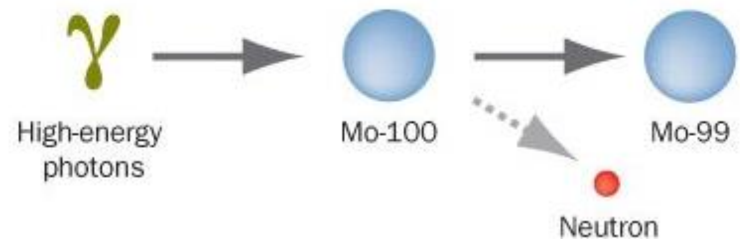
Mo-99 Topical meeting
Washington, DC
June 26, 2014

Proof of Concept Demonstrations for Electron Accelerator Production of ^{99}Mo

- Under the direction of the NNSA, ANL and LANL are partnering with NorthStar Nuclear Medicine, 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 should be commercially available for $400-$600 per gram for kg quantities.



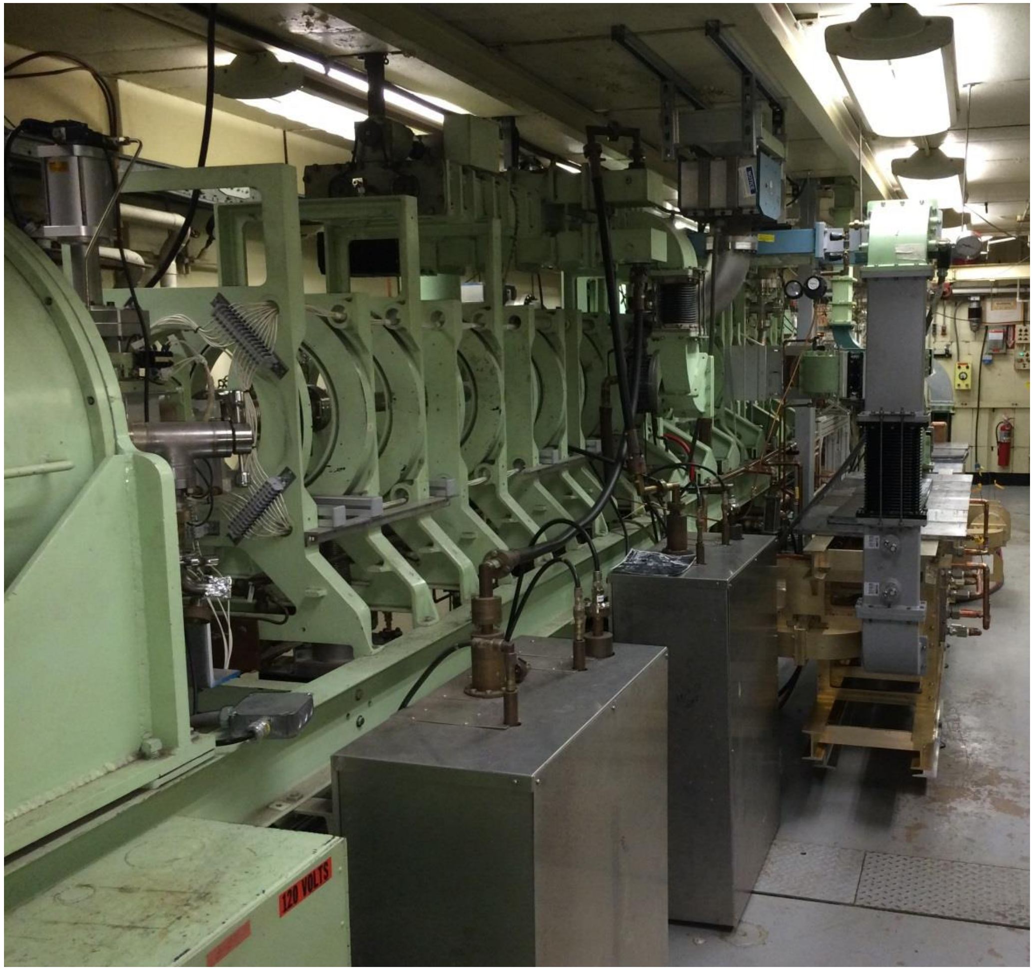


Comparison of the bremsstrahlung photon spectra produced with 20- and 35-MeV electron beams in a Mo target compared with photonuclear cross section of ^{100}Mo.

Scaled Accelerator Tests at Argonne National Laboratory

Seven tests have been performed using the electron accelerator at Argonne.

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.
July 2013	1000-hour He cooling system test
April 2014	Latest thermal test at 35 and 42 MeV with closed-loop He cooling.



Latest Thermal Performance Test
April 2014

- Successfully conducted the thermal test of the 12 mm Mo target and irradiated an instrumented target at 35 and 42 MeV beam energy and power on the target up to 17 kW.

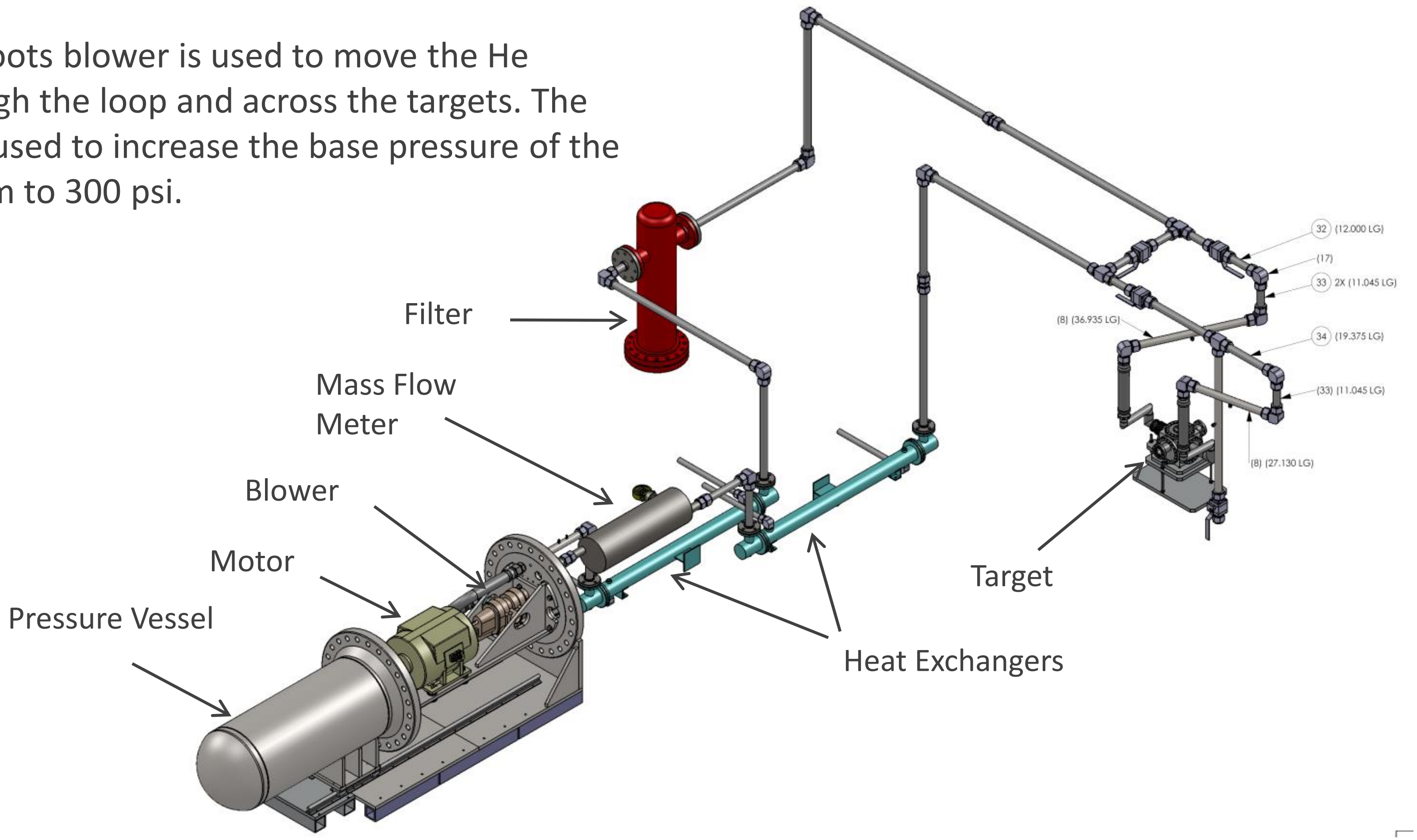
- Thermal data for the target were acquired at different He pressures and flows in the cooling loop. The target performed well.

- Results of the experiment are being analyzed. There are several improvements/issues that have to be addressed.

- Shielding for the OTR and IR cameras has to be improved. There were multiple recoverable communication issues with both the IR and OTR cameras.

Closed Loop Gaseous Helium Cooling System Layout

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.



Future Work (August - October 2014)

Production Test Matrix

	Production Test 1	Production Test 2	Production Test 3	Production Test 4	Thermal Test	Production Test 5
Purpose	Test Enrichment 1 at high energy	Test Enrichment 2 at high energy	Test Enrichment 3 at high energy	Test Enrichment 2 at low energy	Validate the thermal performance of the target	Test Enrichment 4 at high energy for long duration
Energy (MeV)	42	42	42	35	42 and 35	42
Current (uA)	240	240	240	500	300 and 550	240
Power (kW)	21	21	21	17.5	12.6 and 19.3	21
Duration (hours)	24	24	24	24	2	156
Targets	E1 (97.39%) and Natural	E2 (99.03%) and Natural	E3 (95.08%) and Natural	E2 (99.03%) and Natural	Natural	E4 (95.08%) and Natural
Mo-99 EOB Activity [Ci]	5.4	5.3	5.3	9.6	0.2 and 0.28	19.2
Target Thermocouples	No	No	No	No	Yes	No

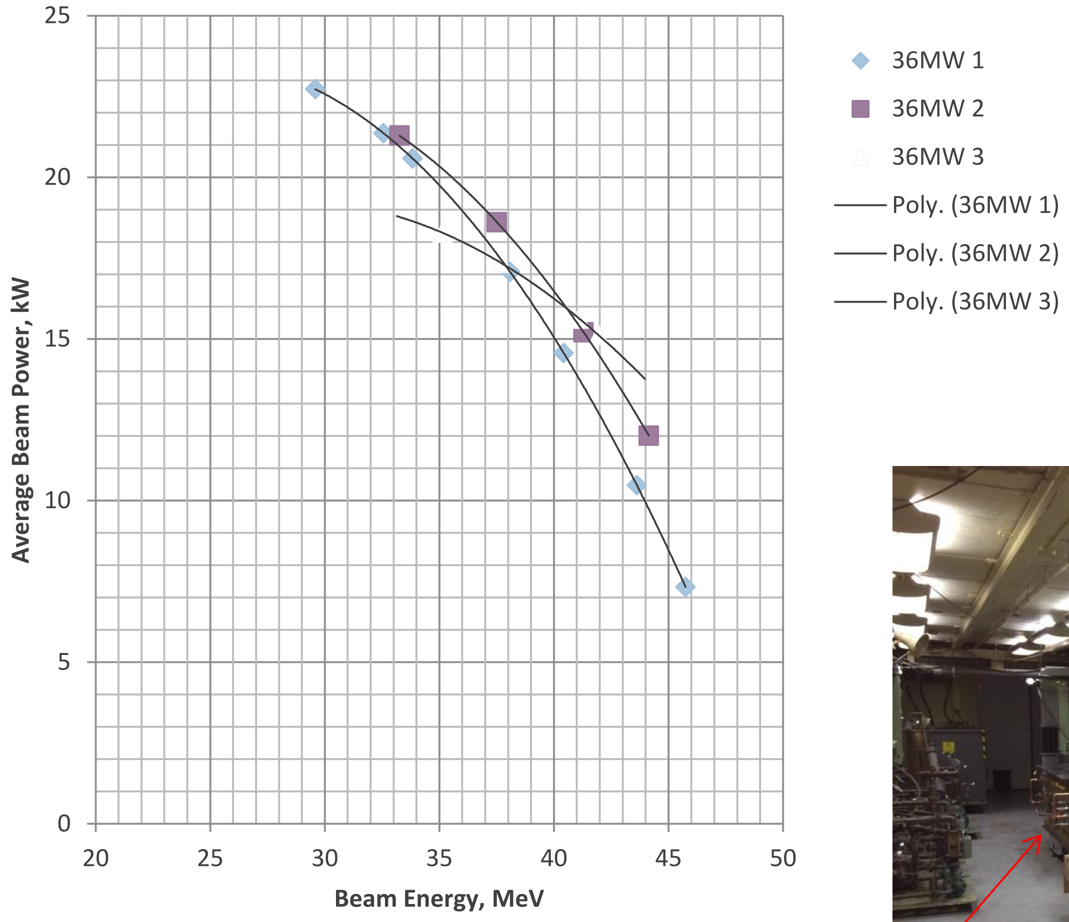
LINAC upgrade

Beam parameters after upgrade (MEVEX proposal)

Energy (MeV)	15	20	25	30	35	40	45	50	55
Beam Peak Current (mA)	1390	1230	1060	900	740	570	390	240	80
Average Beam Current (μA)	1112	984	848	720	592	456	312	192	64
Average beam power on the target (kW)	16.76	19.64	21.32	21.6	20.66	18.28	14.2	9.6	3.6

July 2011	Order for new accelerator structures and circulators was placed
September 2012	Structures arrived
November 2012	Circulators arrived
January 2013	Installation completed, first beam measurements
February 2013	Consultation with MEVEX on low beam-energy
March 2013	RF measurements with MEVEX engineers and repair of circulator 1
April 2013	Second RF measurements. Problem is localized to the circulators being inadequate
June 2013	New circulators are ordered
September 2013	New circulators have arrived
October 2013	New circulators have been installed. Arcing in circulator 1
November 2013	Sent circulator for repair
January 2014	Repaired circulator arrived and installed
February 2014	RF conditioning started
March 2014	Beam tests and start of normal operation

Accelerator performance

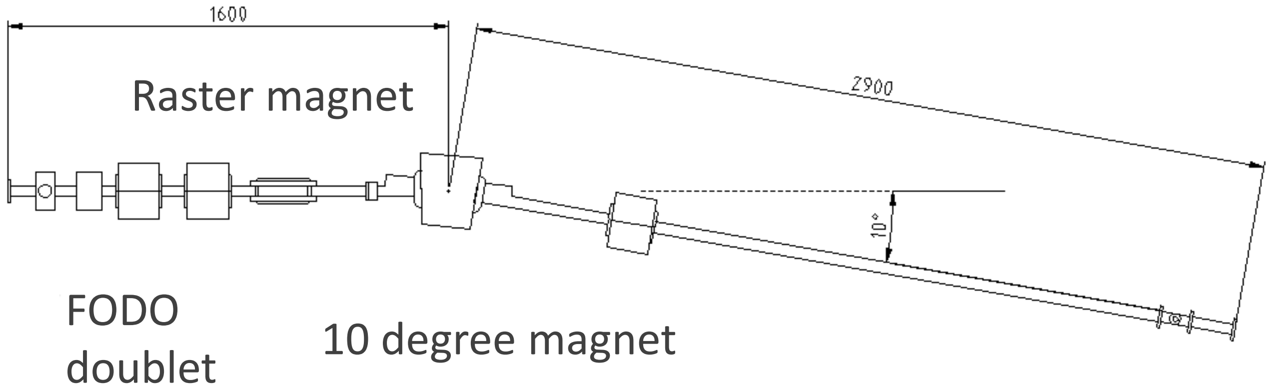


Load lines for upgraded linac

Completely upgraded linac

New RF circulators

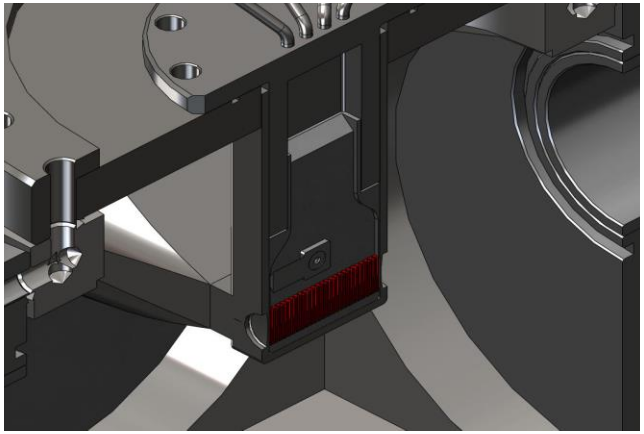
Production facility beam line design



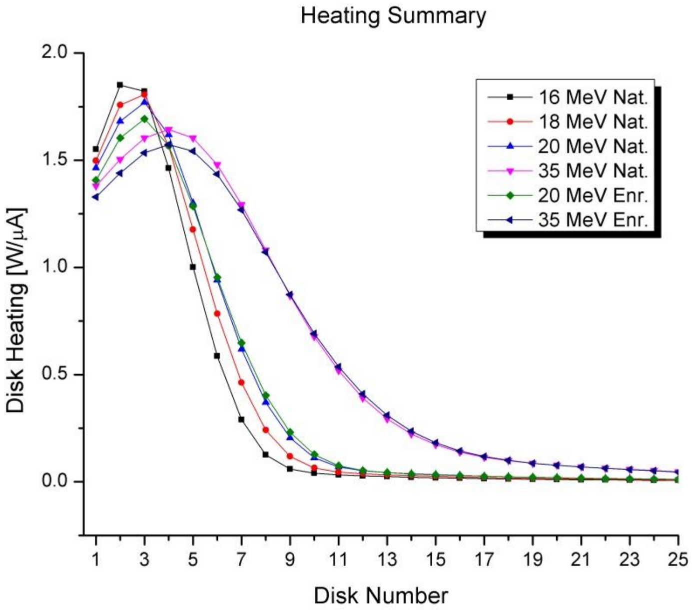
Test beam line at Argonne

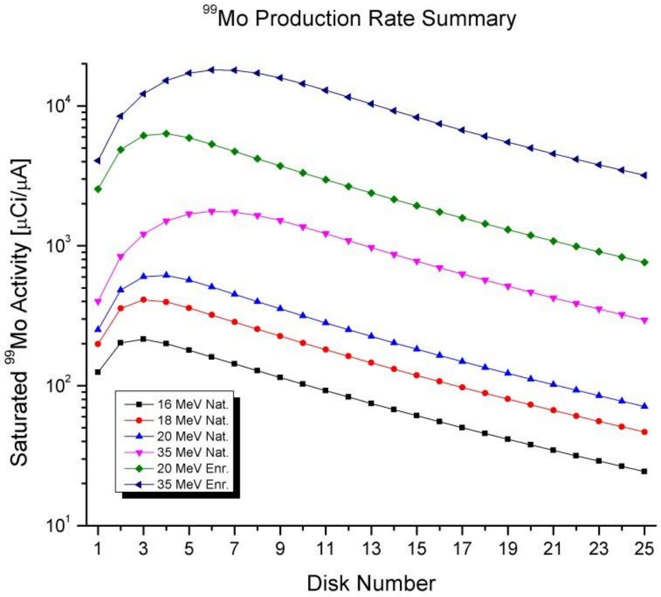
10 degree prototype magnet

MNCPX calculations for Mo-99 production



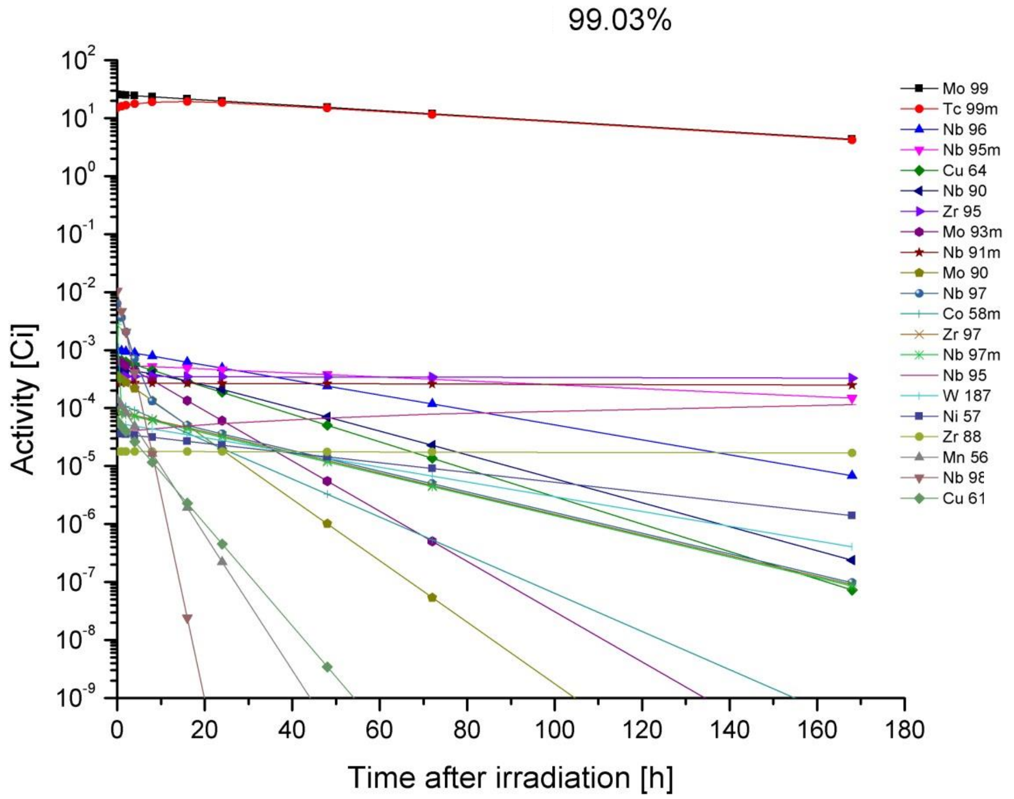
Target:
- 25 disks
- 1 mm thick
- 12 mm diameter





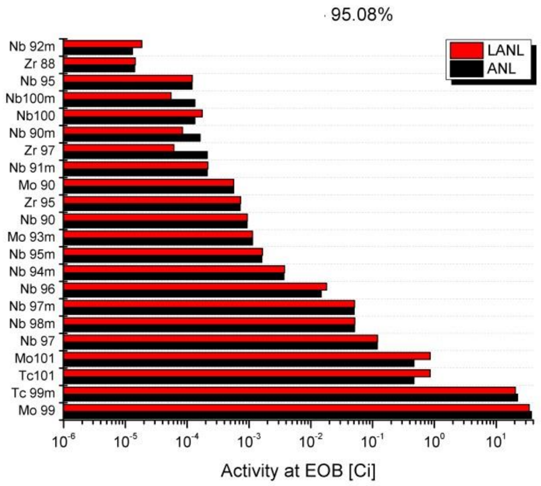
Increase of beam energy decreases peak power in the target and thermal load on the window.

Side-Reaction Modeling of 95.08% Enriched Mo-100 Target

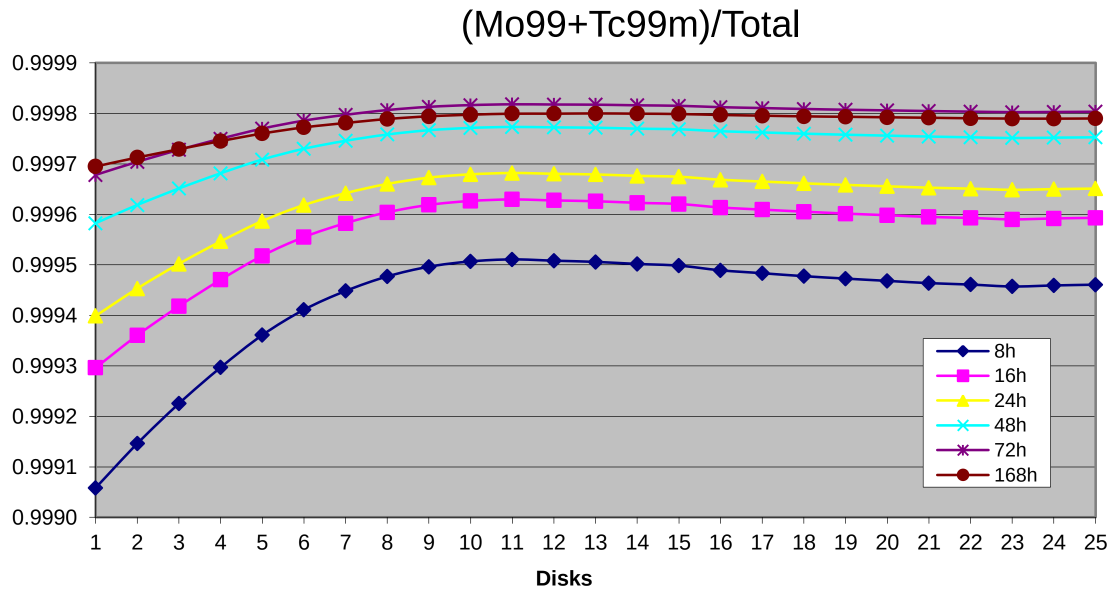


30 MeV 18 kW beam
24 h Irradiation

35 MeV 24.5 kW beam
24 h Irradiation



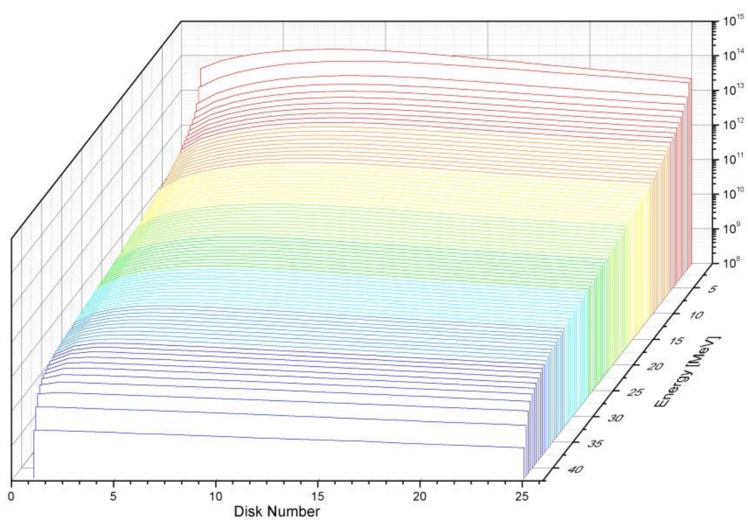
Side-Reaction Modeling at 42 MeV for 95.08 enriched Mo-100

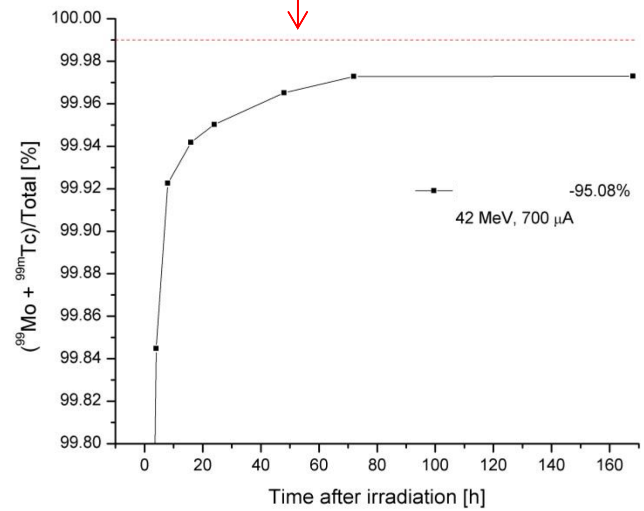


Mo99/Tc99m purity
Disk-by-disk

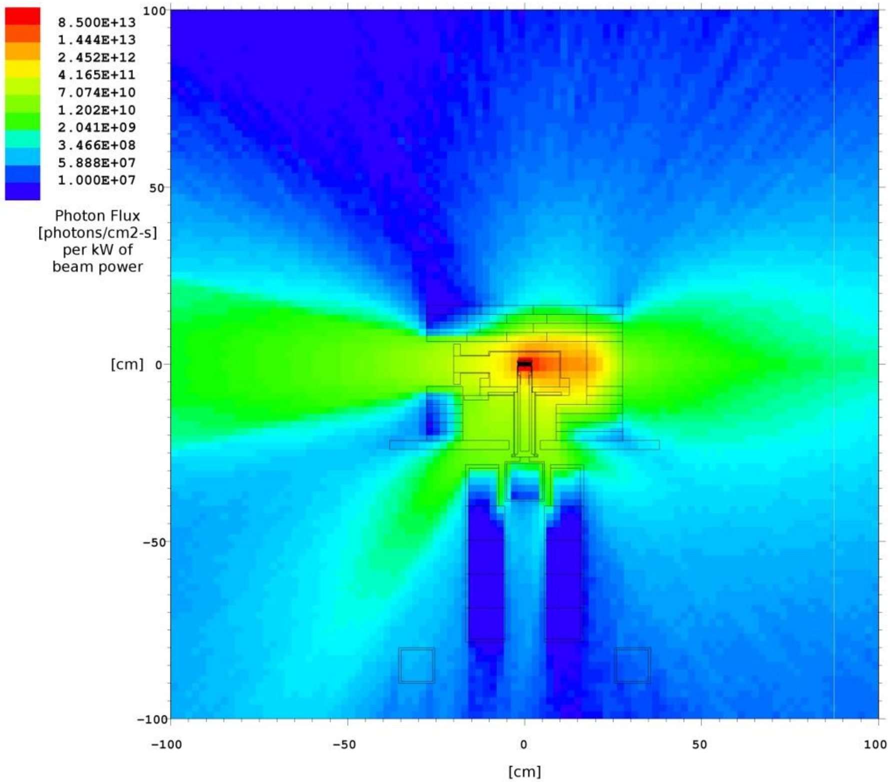
29.4 kW (700 µA)
24 h Irradiation
95.08% Enriched
Mo100 Target

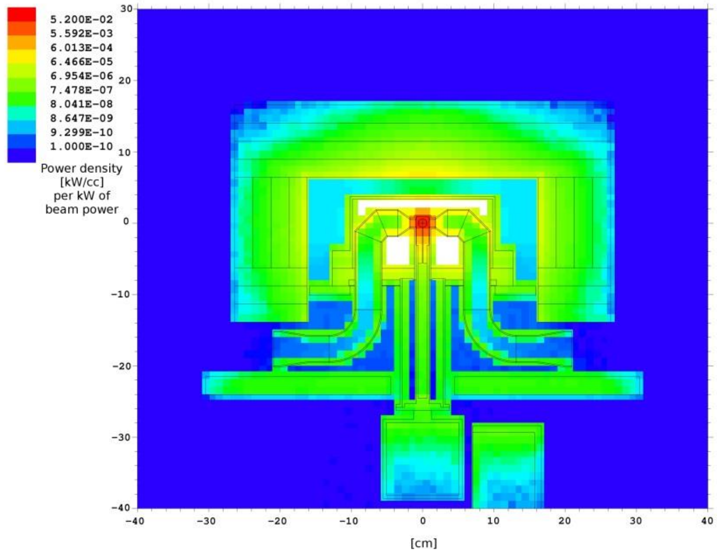
European Pharmacopoeia Requirement

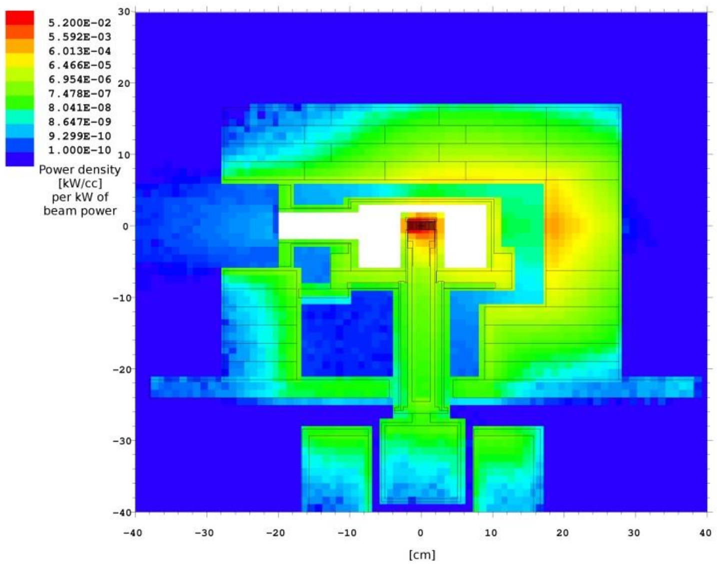




Latest Experimental Design
MCNPX Results







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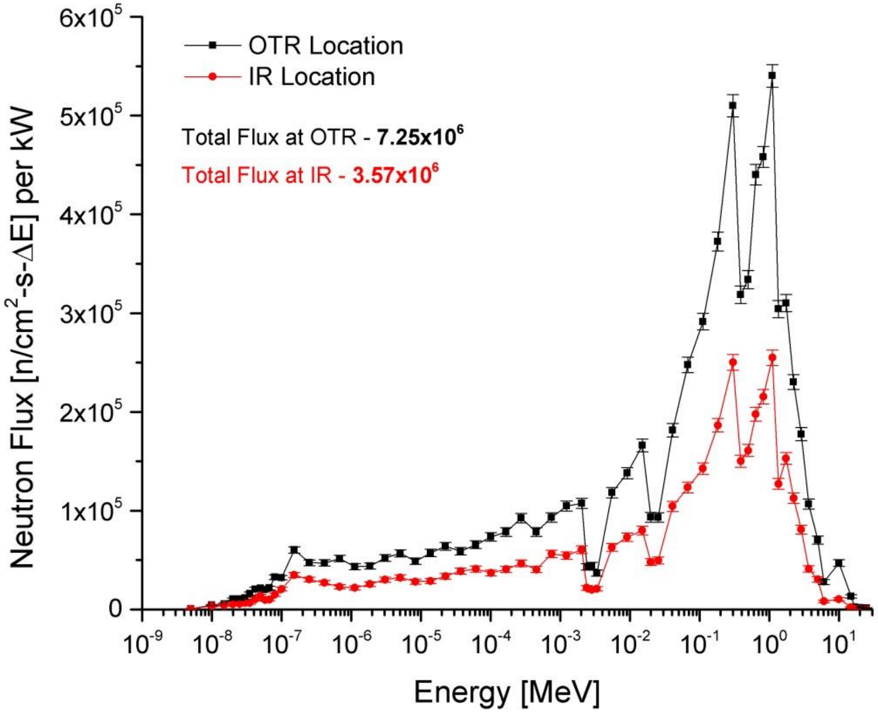
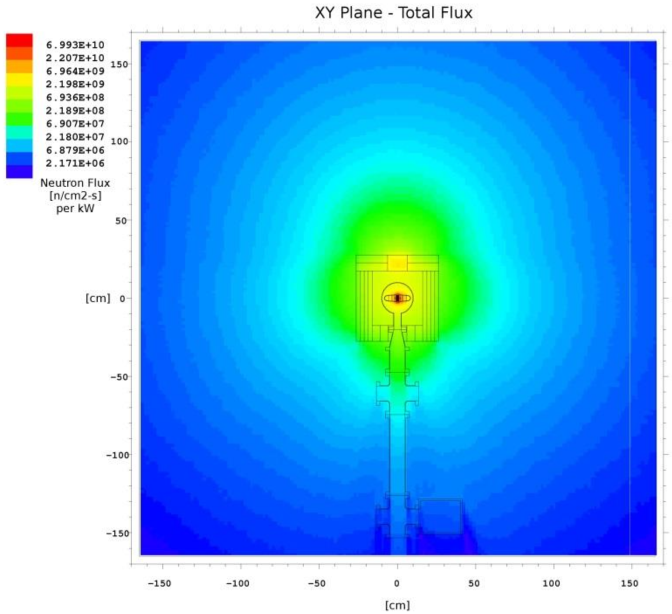
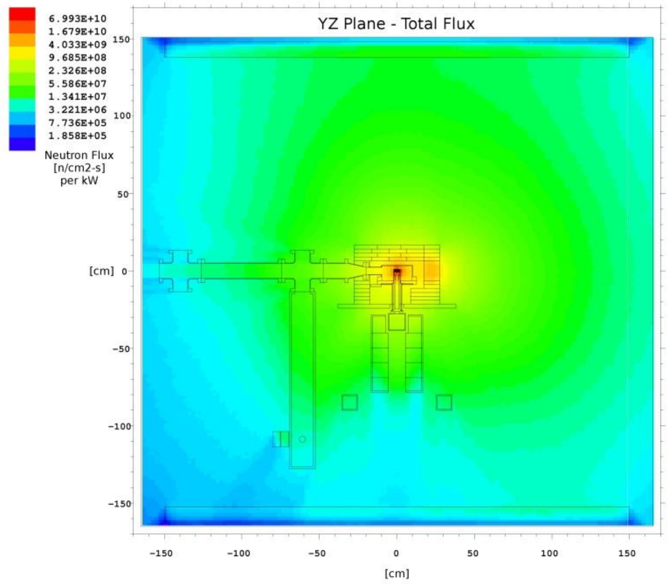
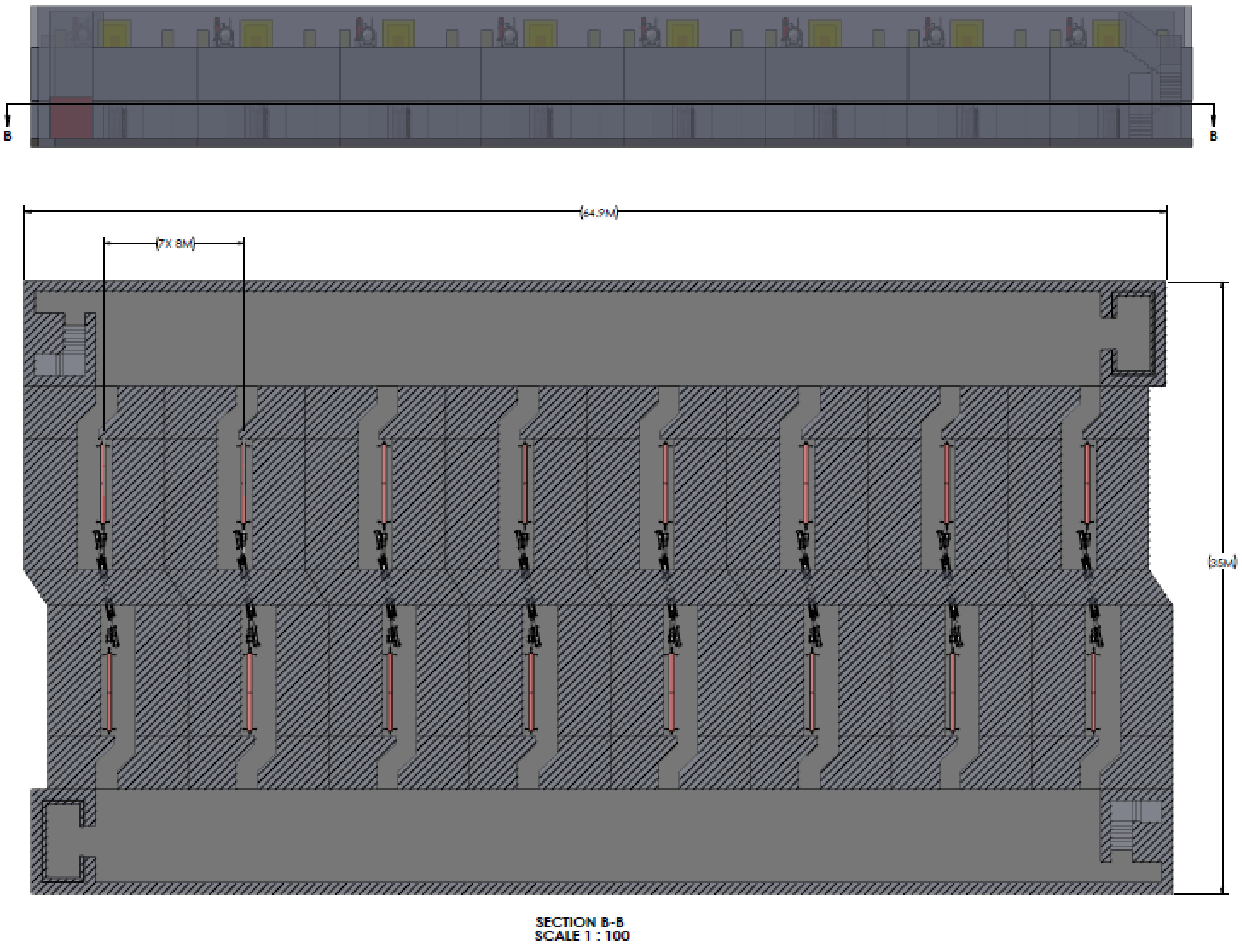


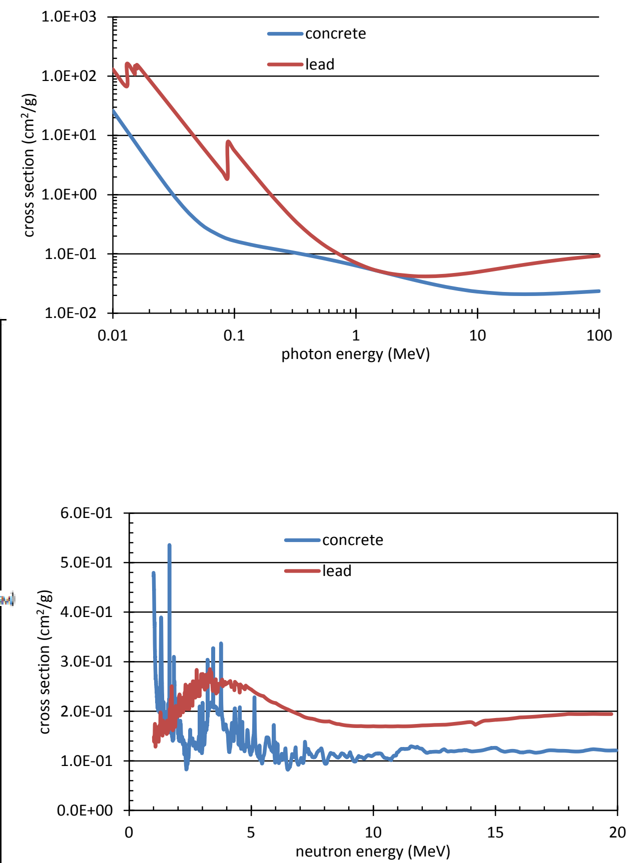




MCNPX Calculations for Production-Facility Shielding

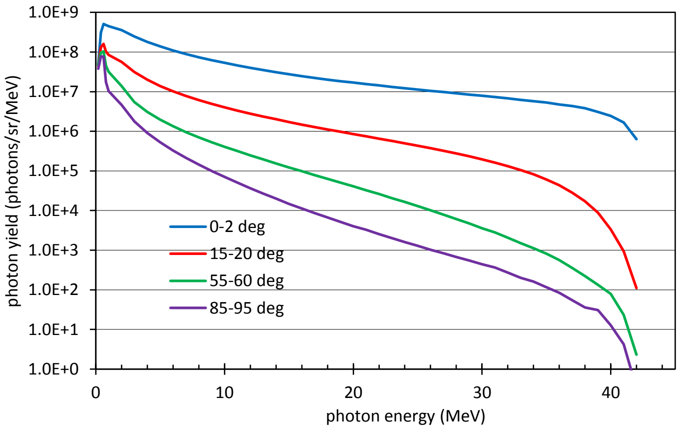


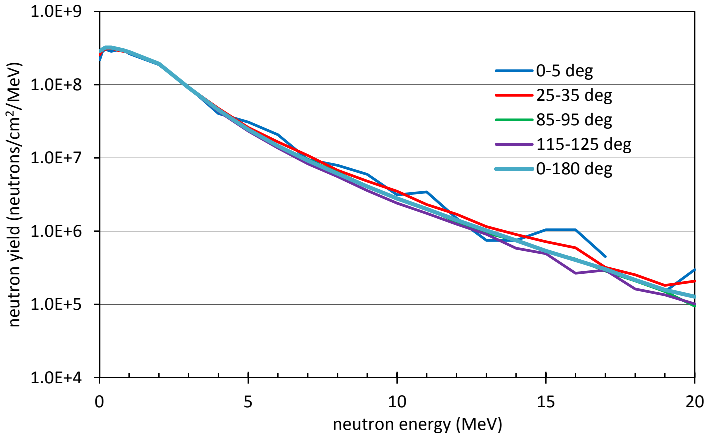
Draft layout of the proposed accelerator facility



Neutron and photon cross sections for lead and concrete

MCNPX Calculations for Production-Facility Shielding





0° emission.

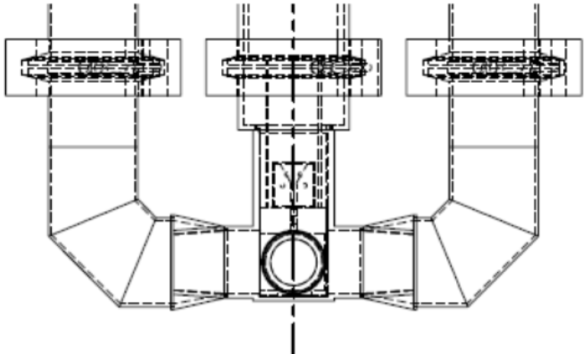
	neutron source		photon source		
concrete thickness (cm)	neutron dose rate (rem/hr)	photon dose rate (rem/hr)	neutron dose rate (rem/hr)	photon dose rate (rem/hr)	total dose rate (rem/hr)
150	3.84e-4	2.87e-3	2.65e-2	2.57e-1	2.87e-1
200	5.34e-6	1.15e-4	3.34e-4	1.02e-2	1.07e-2
250	8.50e-8	4.93e-6	4.56e-6	4.61e-4	4.71e-4

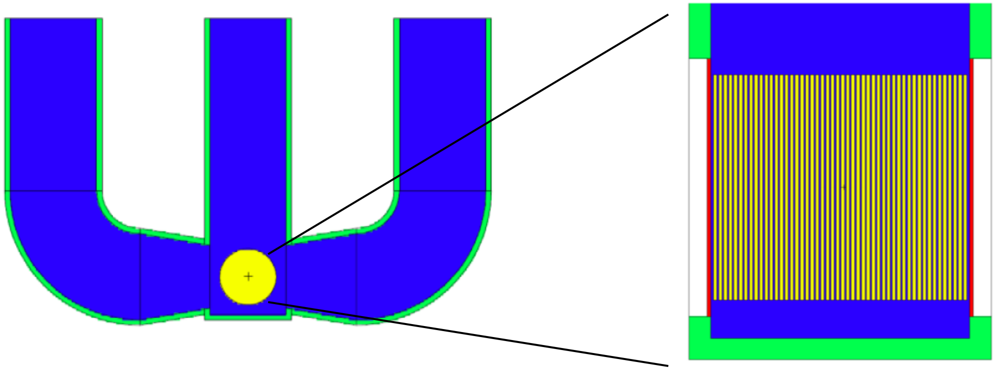
90° emission.

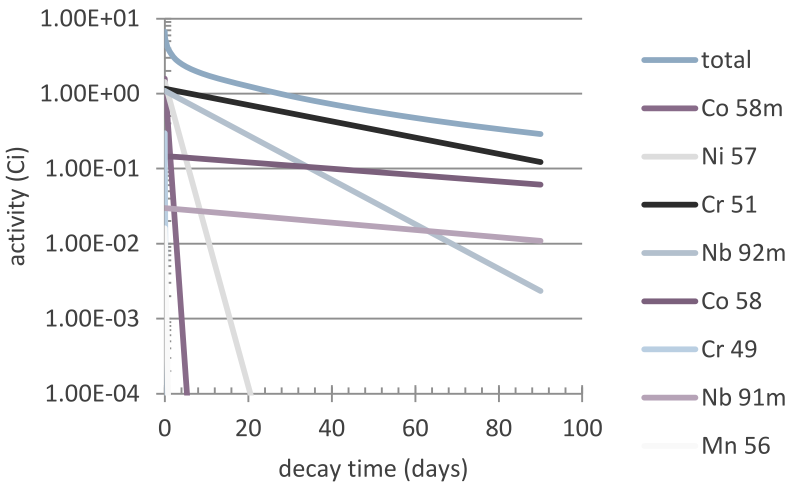
	neutron source		photon source		
concrete thickness (cm)	neutron dose rate (rem/hr)	photon dose rate (rem/hr)	neutron dose rate (rem/hr)	photon dose rate (rem/hr)	total dose rate (rem/hr)
100	8.74e+0	2.27e+1	5.32e-1	1.47e+0	3.34e+1
200	7.78e-4	1.70e-2	3.40e-5	9.49e-4	1.88e-2
250	8.88e-6	6.04e-4	3.42e-7	3.34e-5	6.46e-4

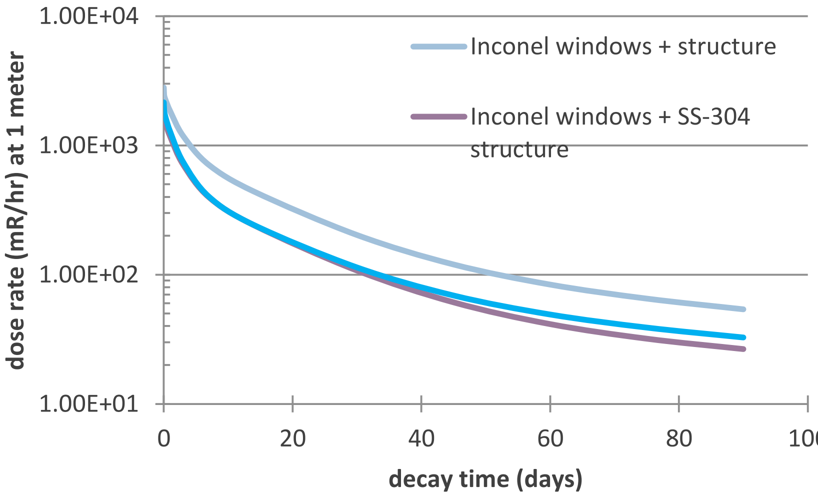
Dose rate for primary and secondary radiations in shield of 30 cm lead + concrete for 120 kW of 42-MeV electrons incident on molybdenum.

Dose Calculations For Production-Target Housing



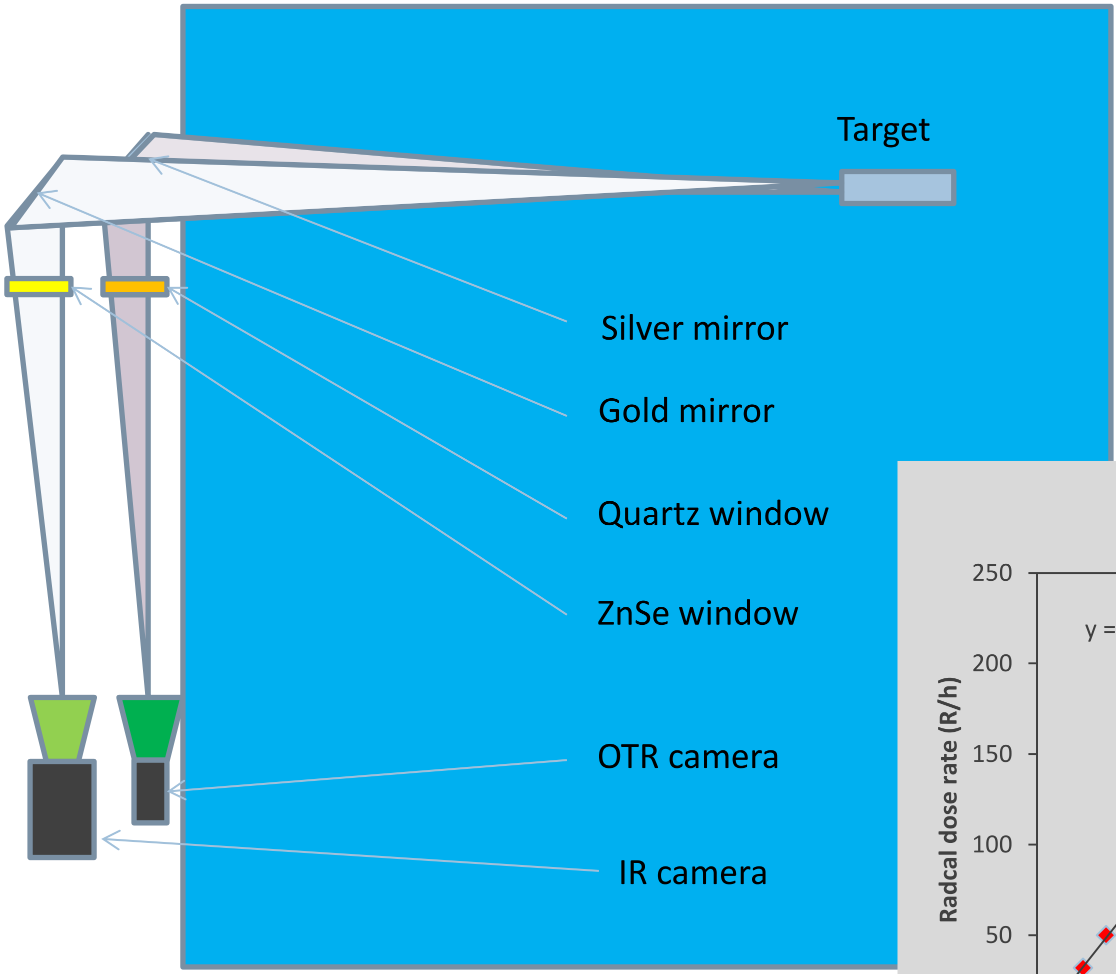


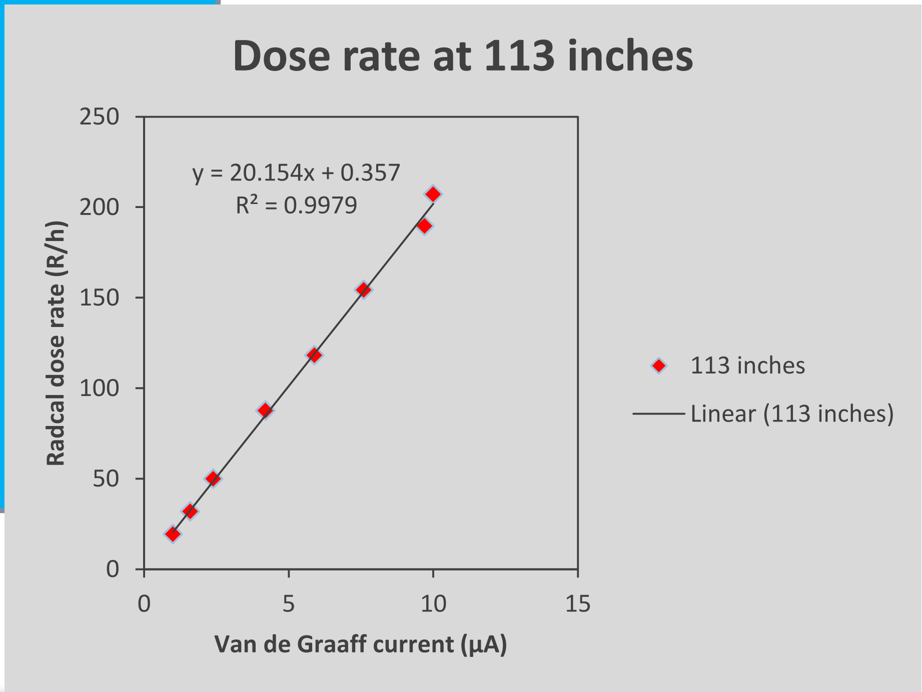




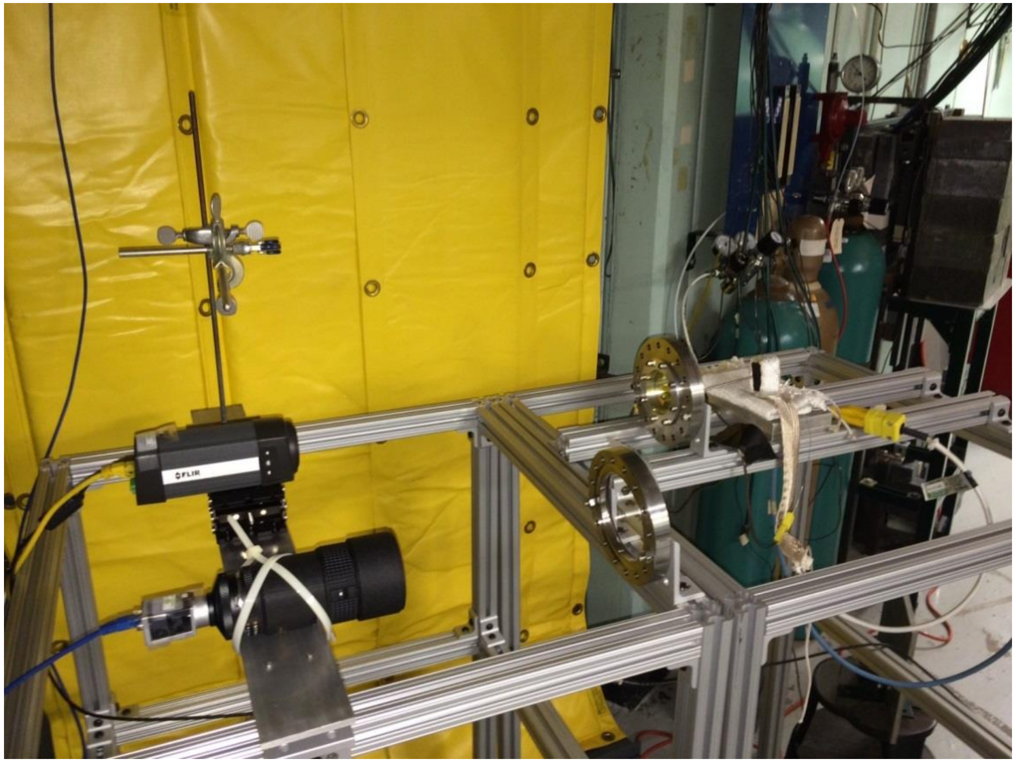
Substitution of the Inconel for stainless steel will reduce dose by the factor of 2

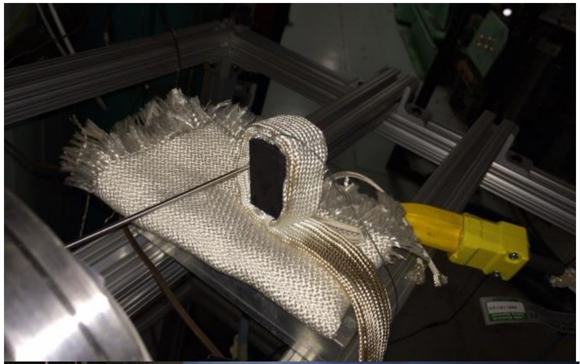
Radiation Testing of Cameras at the Van de Graaff Accelerator Facility





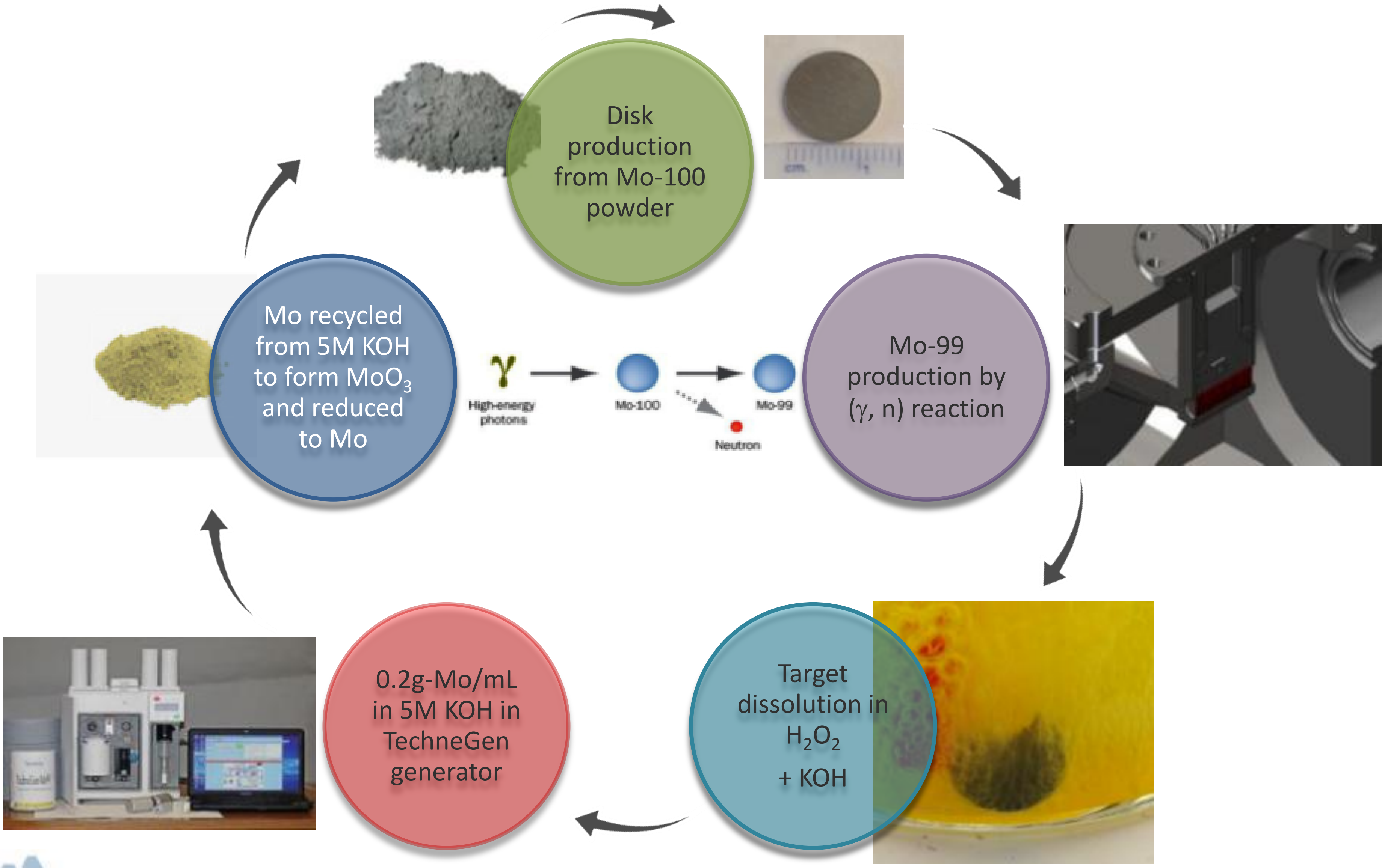
Radiation Testing of the Cameras





Testing at the Van de Graaff accelerator showed that cameras will survive more then a year in the facility

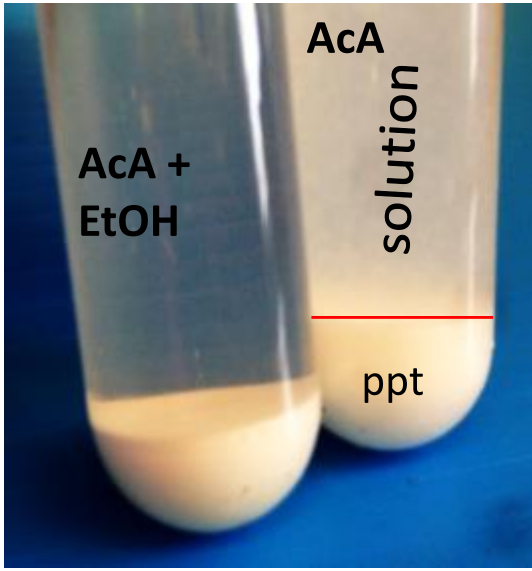
Molybdenum cycle



Mo recovery

1mL of K_2MoO_4 in 5M KOH + 5mL of reagent (**1:5 ratio**)

Reagent	Mo lost	K removed
Glacial AcA	0.2-2%	70-80%
70% HNO_3	5-20%	80-90%
Ethanol	0-0.2%	~40%
AcA+ethanol (1:4)	0-0.2%	~40%
H_2SO_4	N/A	N/A



H_2SO_4 - not suitable for Mo precipitation – forms Mo suspension

HNO_3 - not suitable for Mo precipitation – significant Mo loss

Ethanol - not suitable for Mo precipitation – does not remove K from K_2MoO_4

Acetic acid – the best reagent – good removal of K, good Mo recovery

Summary of the Mo recycle and future plans

- Mo can be precipitated from highly alkaline solution using glacial acetic acid
- Mo precipitate is then washed with 70% HNO_3
- Good Mo recovery **97-100%** obtained if 1st HNO_3 wash allowed to sit for several hours
- Purification of potassium **<25 ppm** (99.999% removed) – for small scale, work continues with large scale experiments
- XRD characterization of Mo precipitate – converting to MoO_3
- Large scale experiments look promising and able to process up to 400g of Mo
- HNO_3 can be recycled
- Large scale experiments continue with dissolved irradiated targets
- Precipitation step and washing steps need to be optimized for better Mo recovery

Summary

- We have conducted several irradiation that demonstrated satisfactory target performance. Next tests will be focused on production of Mo-99.
- Simple beam-line design for production facility was developed and tested.
- 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.
- Substitution of Inconel by stainless steel in the target housing will reduce dose by factor of two.
- Cameras testing at the Van de Graaff facility demonstrated sufficient radiation resistance of the equipment.
- Mo recycle process was demonstrated with good efficiency.

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