Development Activities in Support of Accelerator Production of ^{99}Mo Production through the γ /n reaction on ^{100}Mo

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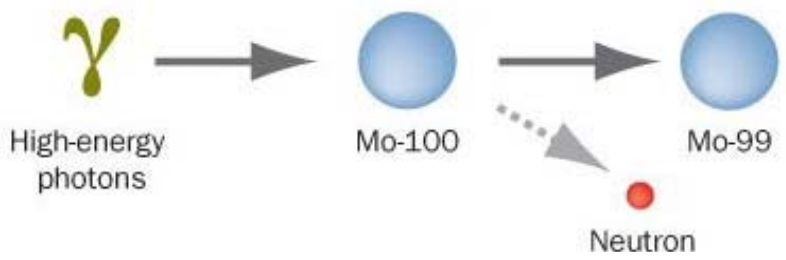
Outline

- **LINAC upgrade and upcoming thermal and production test**
  - MCNPX simulations
  - Testing of He cooling loop
  - Production of several Ci of ^{99}Mo
- **Processing of Mo sintered disks (12 × 1 mm)**
  - Dissolution studies vs. disk properties
  - Dissolving undissolved Mo disk residues
  - Conversion of Mo/H_2O_2 species into MoO_4^{2-}
- **NorthStar generator for separation of $^{99}Mo/^{99m}Tc$**
  - Optimizing the recovery and purity of the product
- **Future plans**

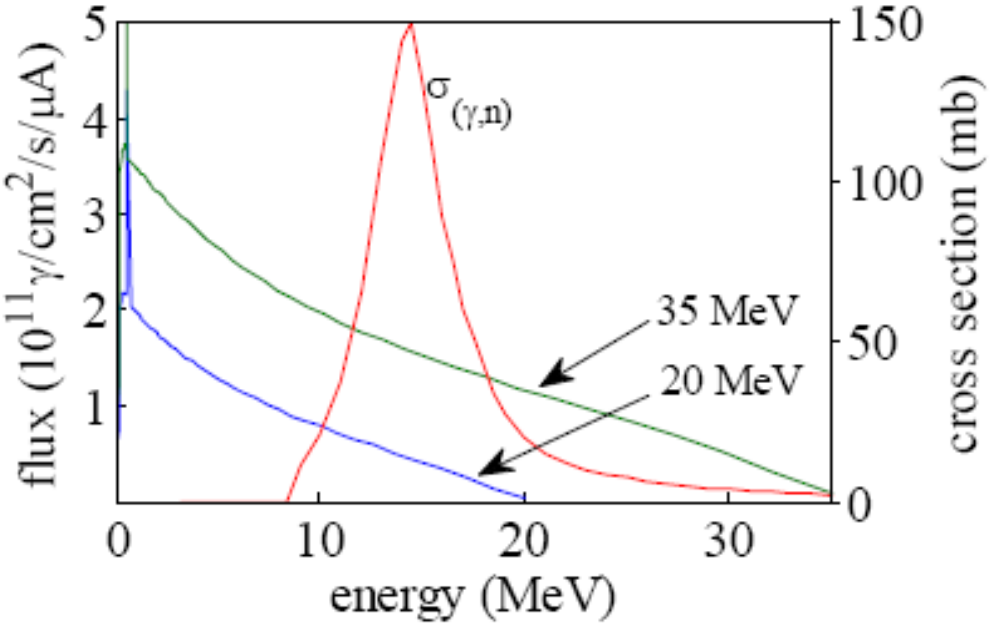
Development of the Accelerator Based Production of ^{99}Mo

- ANL and LANL are assisting NorthStar in development of accelerator based production of ^{99}Mo through the ^{100}Mo(γ,n)^{99}Mo reaction.
- Enriched ^{100}Mo is commercially available for $400-$600 per gram for kg quantities.
- High energy photons are created from a high power electron beam through bremsstrahlung.



$$Y_{m,E_0} = N_m \int_{E_{th}}^{E_0} \sigma_m(\gamma,n)\Phi_{E_0}(E)dE$$

N_m – atom density of material m
E_{th} – threshold energy in m
$\Phi_{E0}(E)$ – photon fluence spectrum
$\sigma(\gamma,n)$ – cross section



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

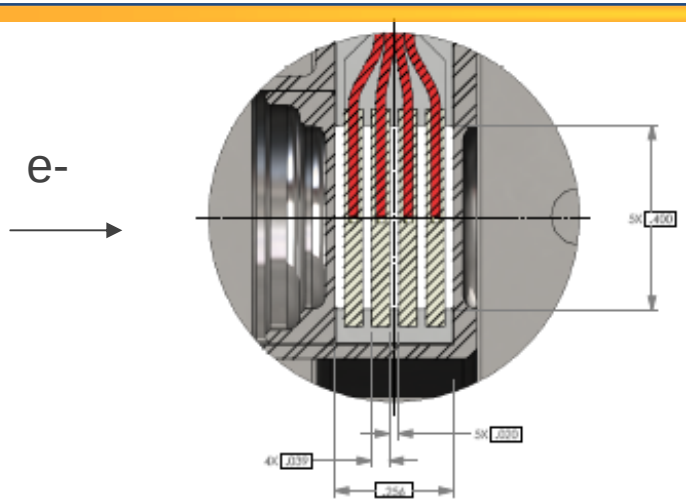
First series Accelerator Tests

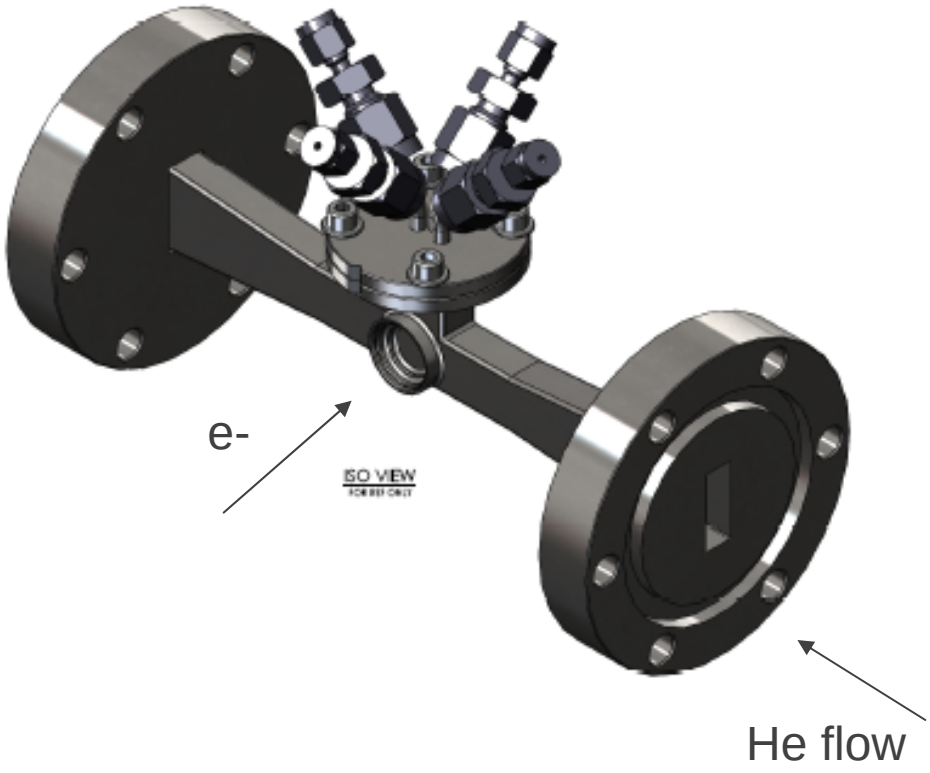
To test and validate the accelerator production process, including the performance of the NorthStar generator, scaled production tests were performed at the Low Energy Accelerator Facility at Argonne National Laboratory (ANL)

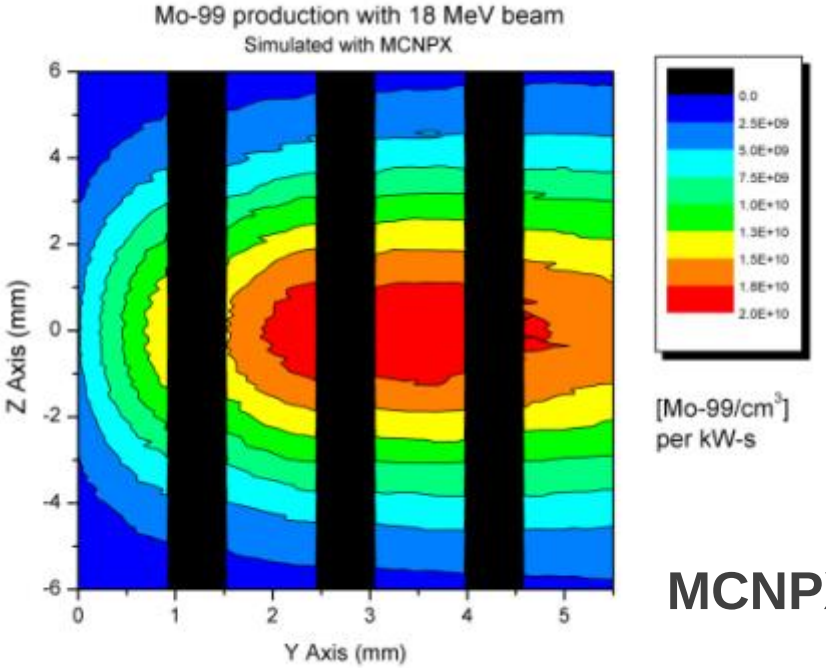
Three low power scaled production tests were completed in 2010 using the ANL electron linac under the following nominal parameters:

2 natural and 1 enriched ^{100}Mo targets (0.23mCi, 0.36mCi, 10.5mCi of Mo-99)
20 MeV, 100 μA, 2 kW beam
6 mm diameter target
6 mm FWHM beam
50 μA on target (1 kW)

Test with 12 × 1mm Mo sintered disks (April 2011)



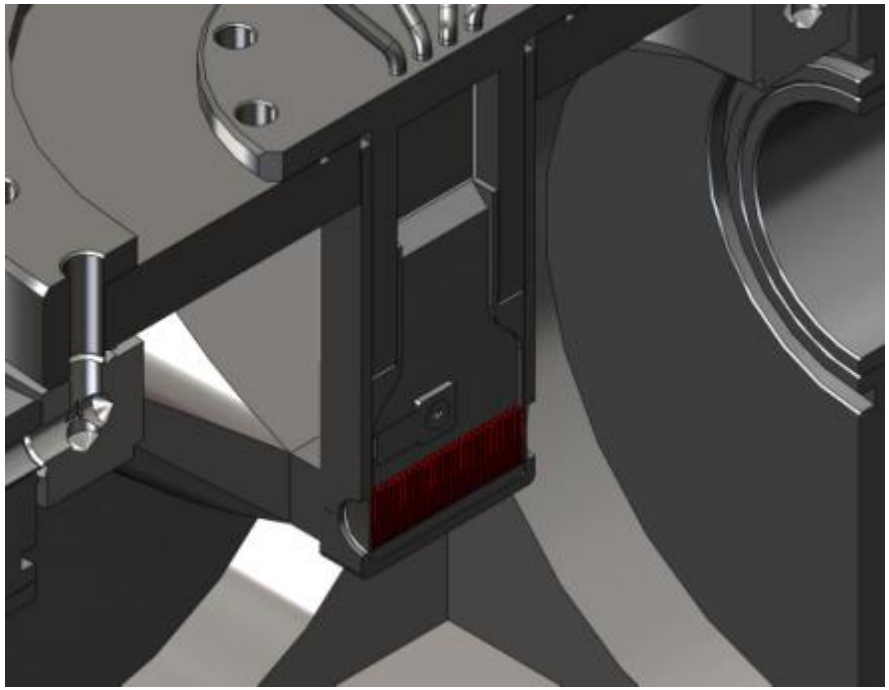




MCNPX simulations April 2011

Upcoming NorthStar experiment – Spring 2012

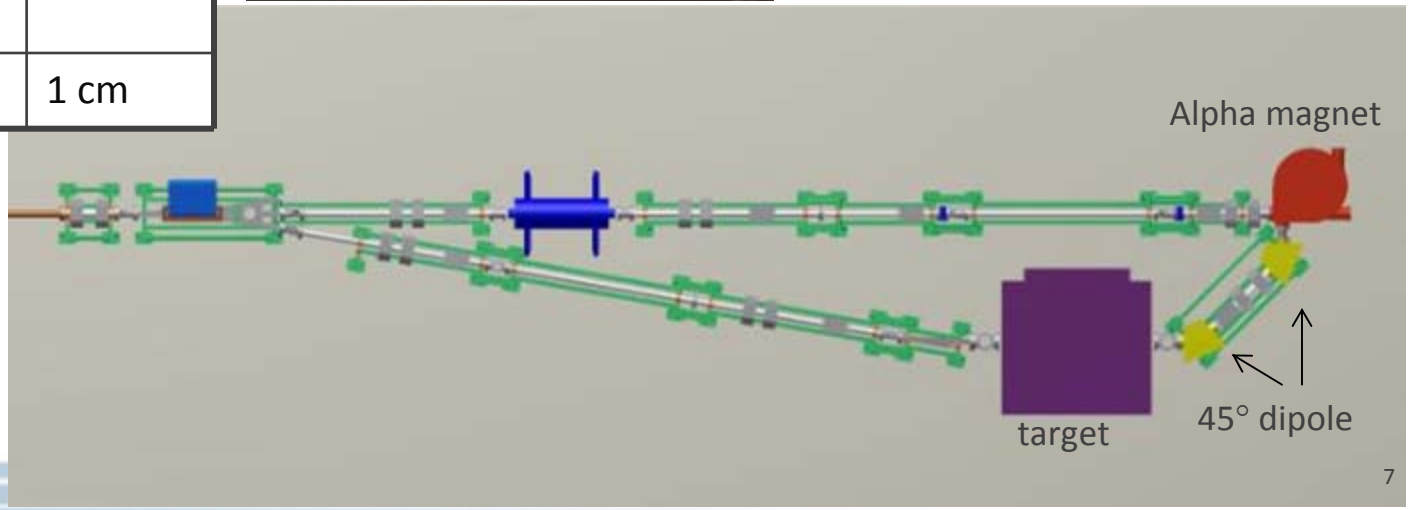
25 Mo disks: 1mm thick 12 mm diameter
Closed loop He flow system
Two test: thermal test with natural Mo disks
production test with enriched Mo-100 disks



Electron Linac upgrade

	Present	Upgrade
Maximum beam energy	21 MeV	58 MeV
Maximum efficiency energy	13 MeV	30 MeV
Maximum beam power	30 kW	30 kW
Pulse width	5.5 μs	4.5 μs
Injector pulse current	2.5 A	1.2 A
Beam size	1 cm	1 cm

- Decrease activation of the equipment in the room
- Allows to extract and transfer sample for analysis and processing
- Reduces dose to personnel



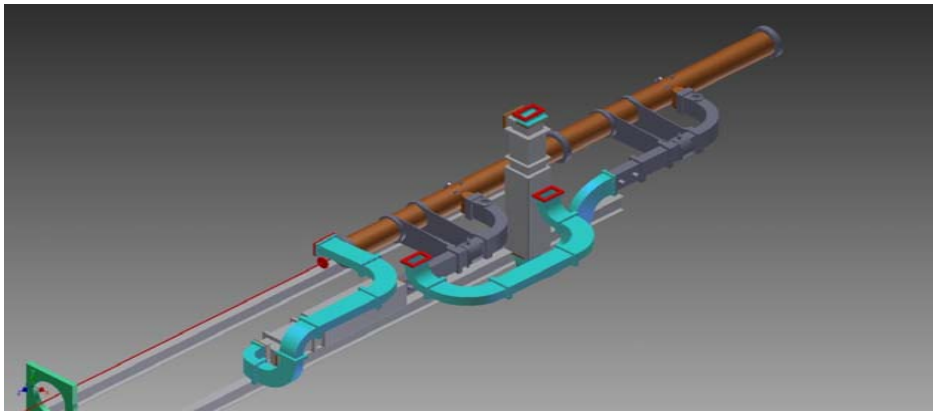
Electron Linac upgrade

Linac beam parameters after upgrade

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

Range of the parameters for: Thermal target testing | Side reactions study | Production runs

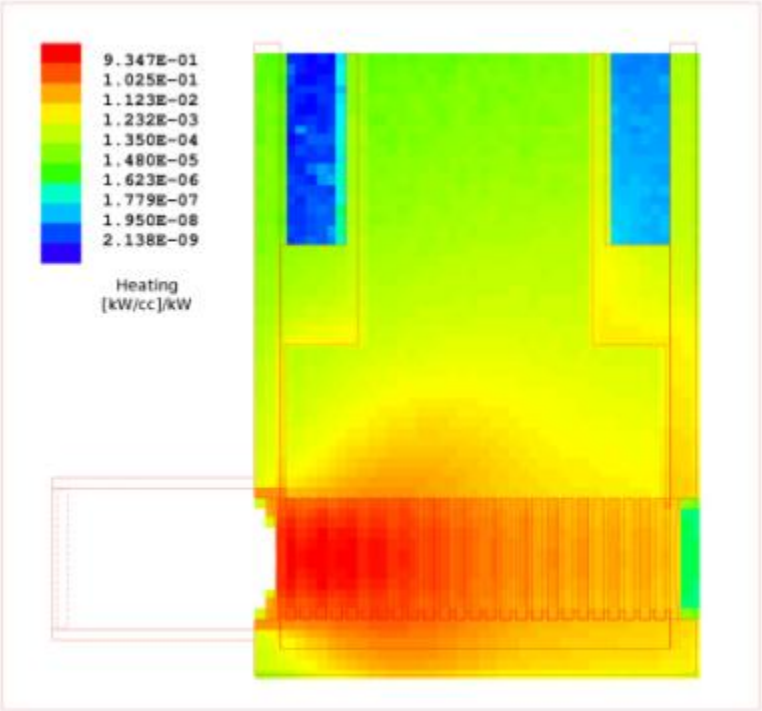
New accelerator structures were delivered to ANL in November 2011



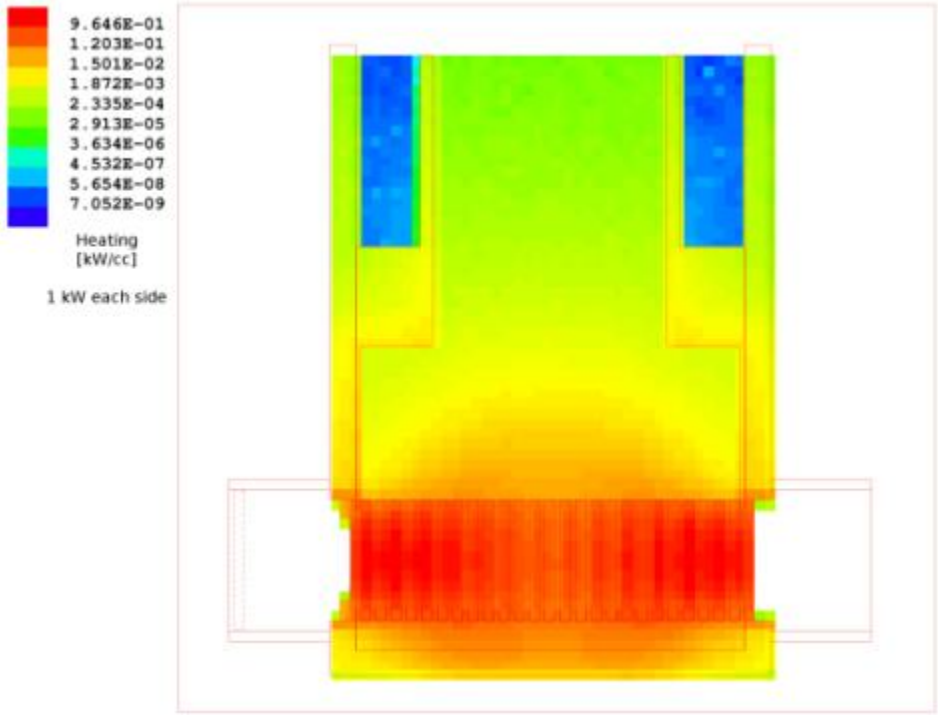
We have developed new RF layout to accommodate new accelerator structures

Heat deposition calculations

Power density distribution [kW/cm^3] per kW of beam power for a 35 MeV beam and natural molybdenum disks for one sided and two sided irradiation



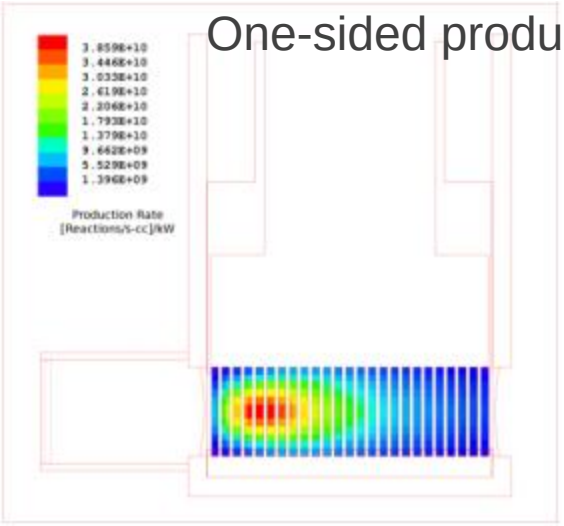
One-sided heating



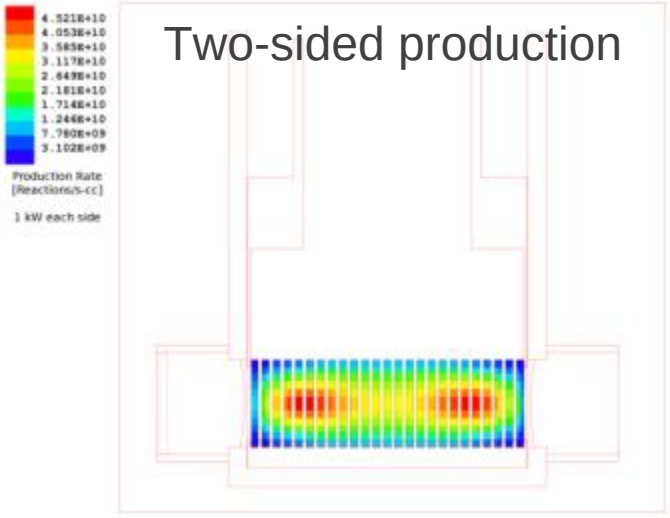
Two-sided heating

Mo-99 production calculations

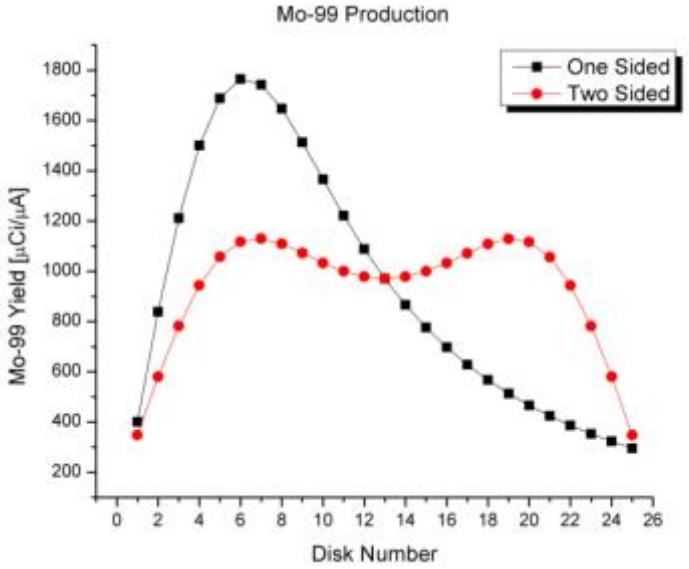
One-sided production

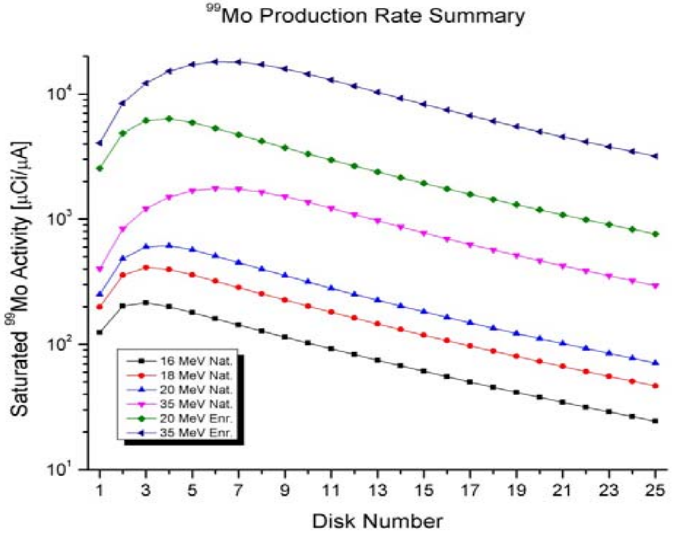


Two-sided production

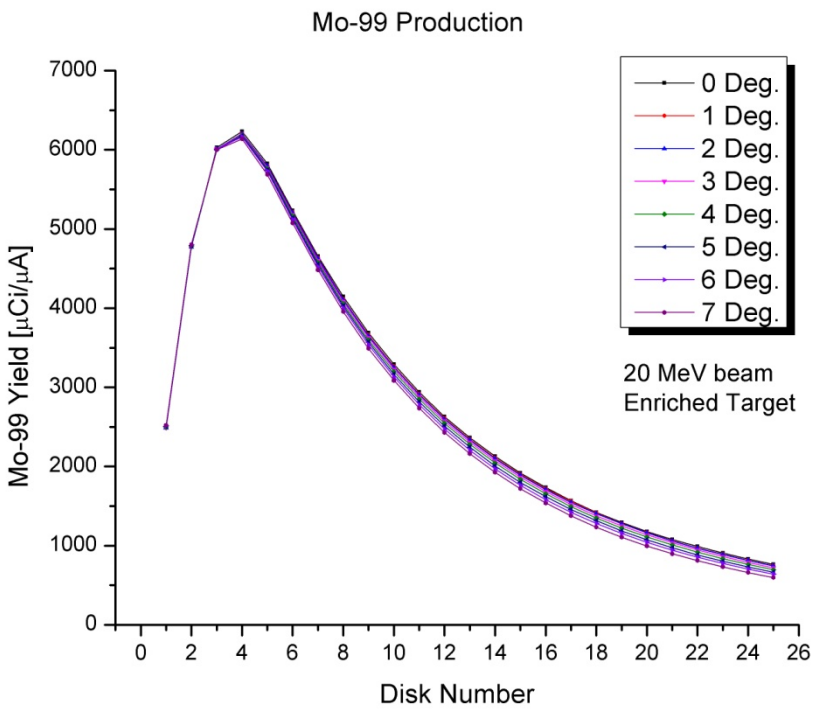


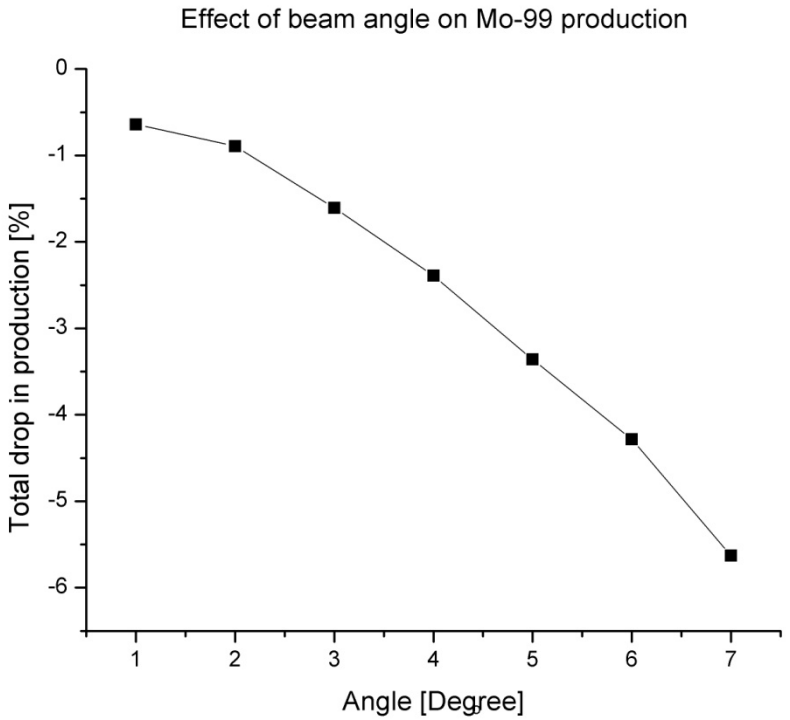
35 MeV beam, Natural Mo disks



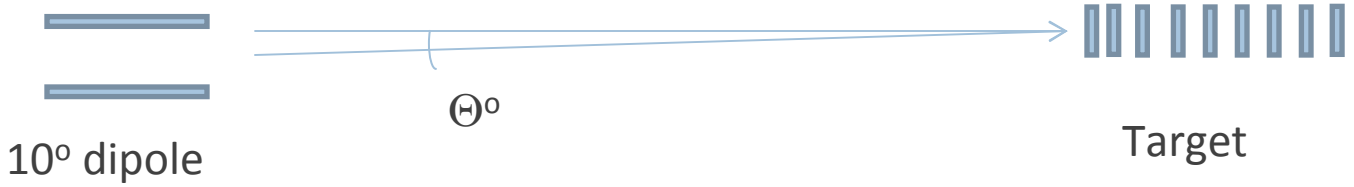


Effect of misalignment





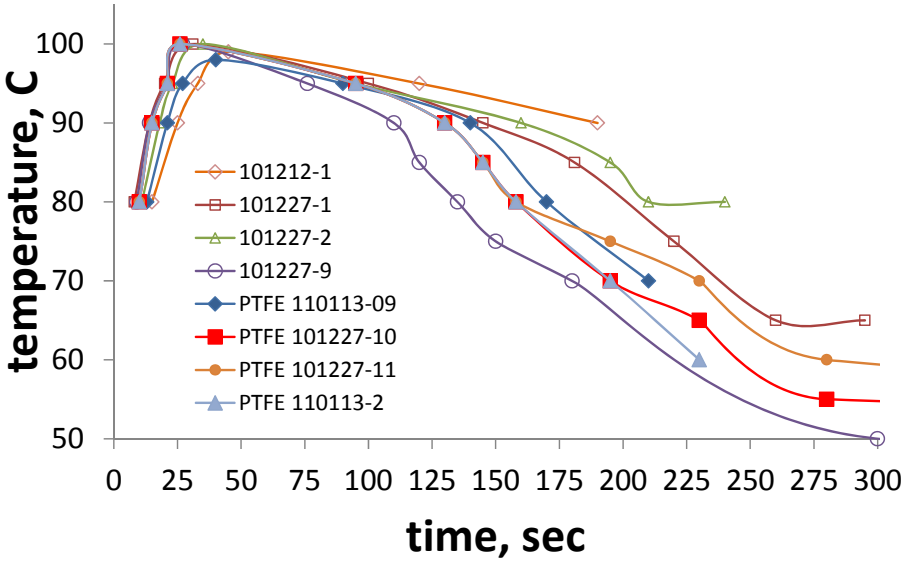
An effect of beam angle on total Mo-99 production. A 20 MeV and enriched target case, simulated with MCNPX

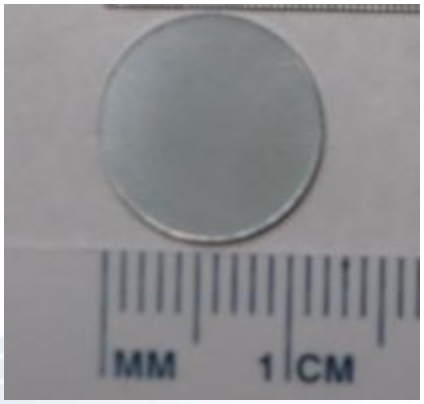


Beam alignment on the target using OTR

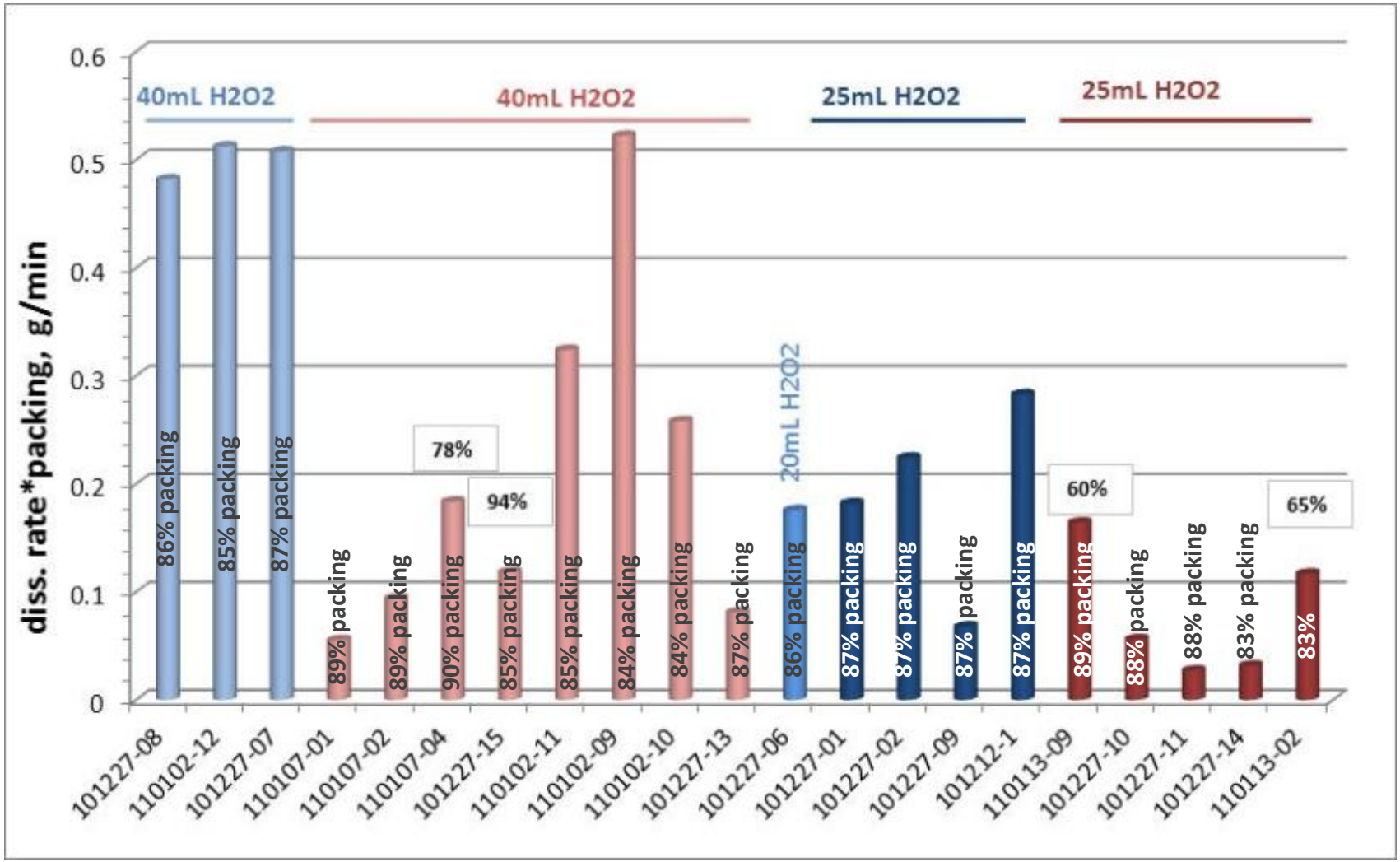
Dissolution of Mo sintered disks in 30% H_2O_2

- $2Mo(s) + 10H_2O_2 = [Mo_2O_3(O_2)_4(H_2O)_2]^{2-} + 2H_3O^+ + 5H_2O$
- Initial pH≈5, after dissolution pH ≈1-2
- 1 Mo disk ~1.07g, 40mL of 30% H_2O_2 at 70°C, H_2O_2:Mo mol ratio ~35

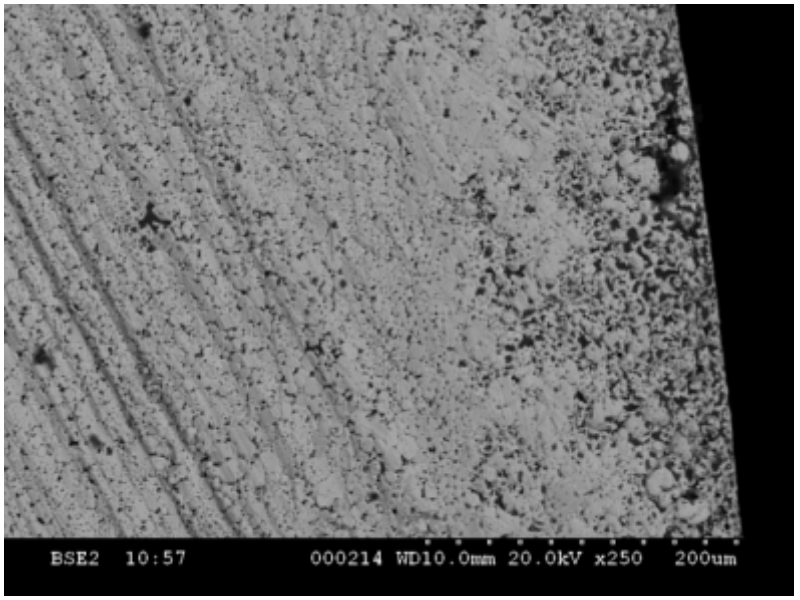


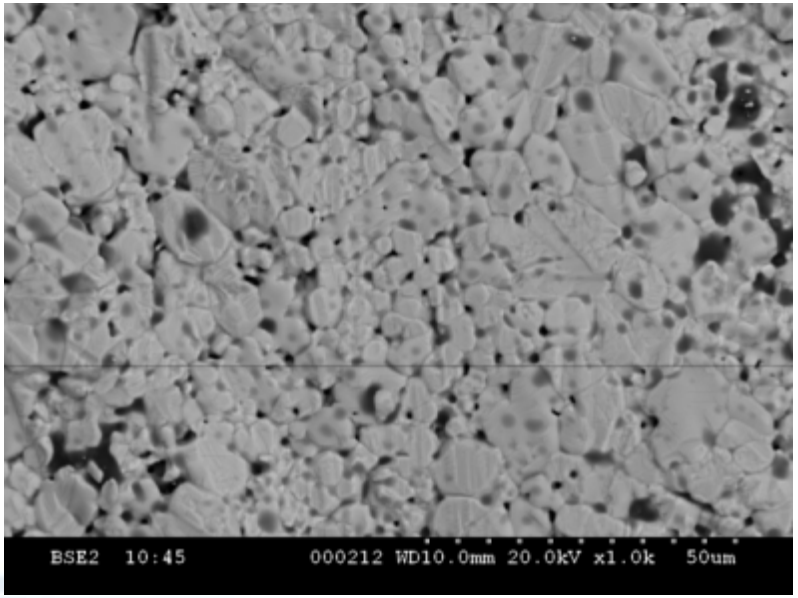


Dissolution rates for some PTFE and non-PTFE disks



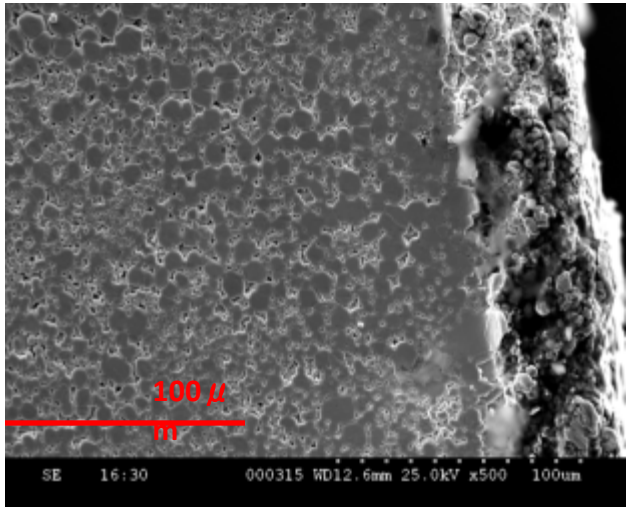
Dissolution of Mo sintered disks in 30% H_2O_2

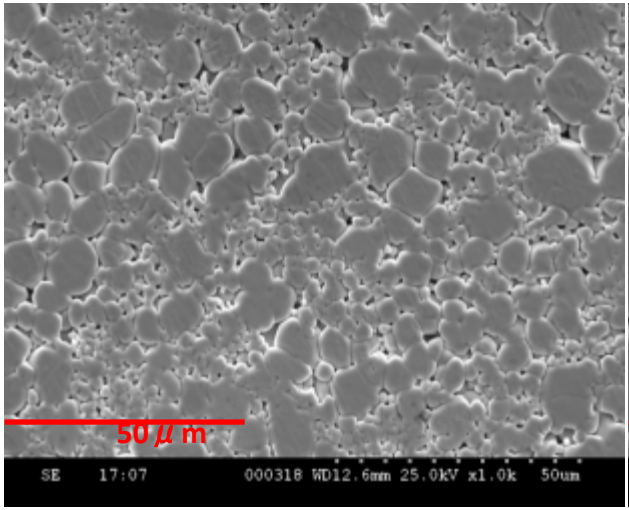


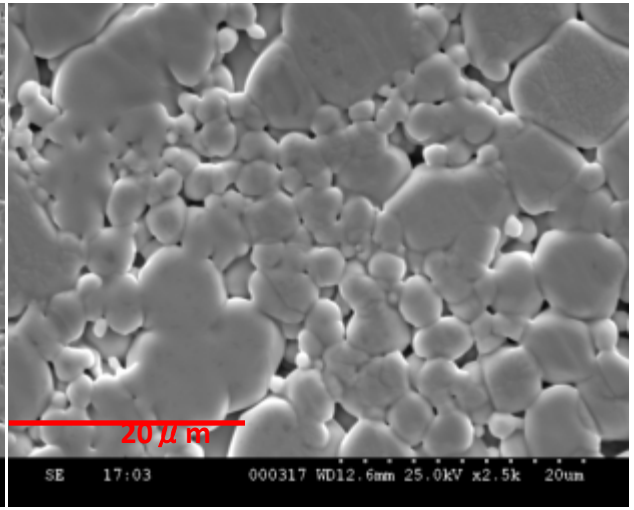


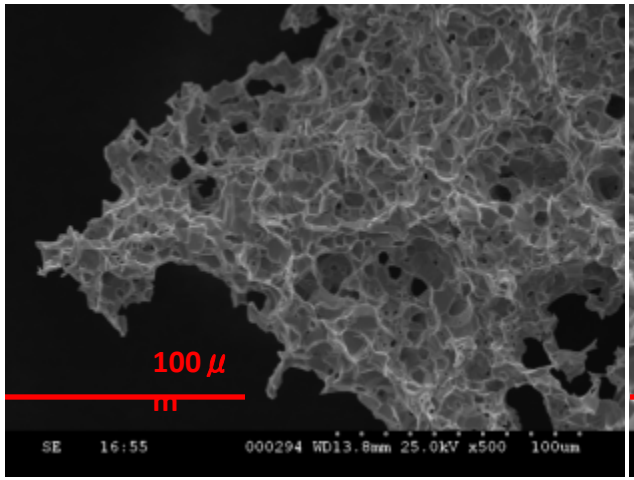
- XRD showed the presence of mostly Mo
- Formation of a thin layer of Mo oxide does not affect the dissolution rate

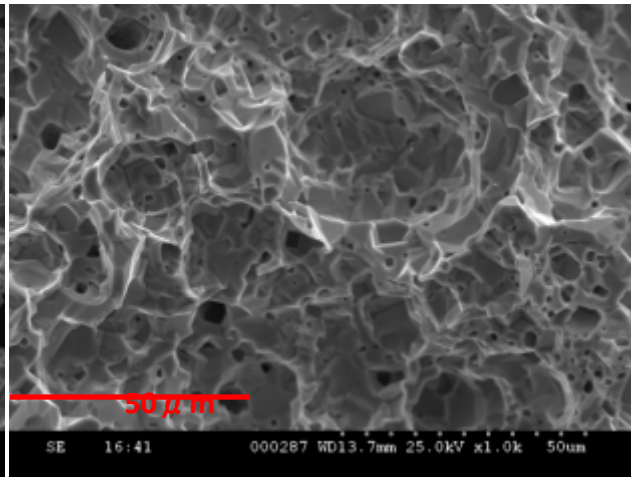
Scanning Electron Microscopy images of Mo disks

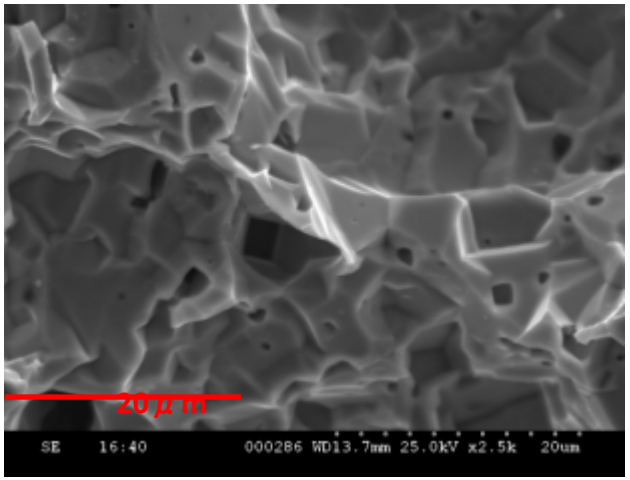






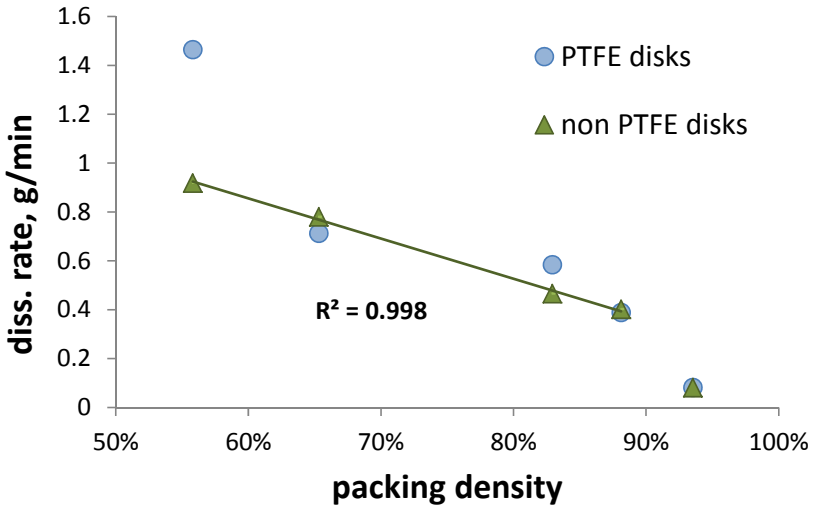






Dissolution rate vs. packing density

New batch of Mo disks – no difference in dissolution rates for PTFE and non-PTFE disks



- High surface area of un-dissolved Mo disk residue
- Surface tension of H_2O_2
- Increase of temperature decreases surface tension
- High packing density disks fully dissolved at ~95-100°C H_2O_2 in about 13-15 minutes

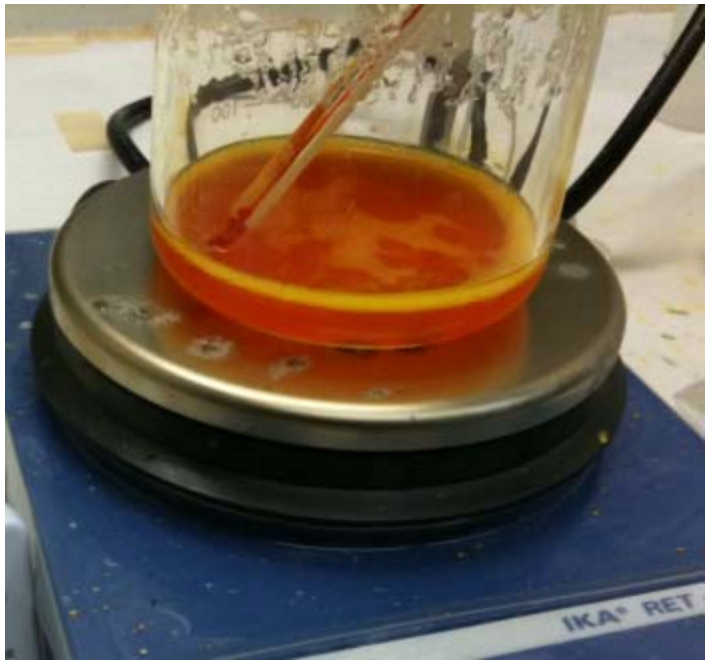
	light sintered disks			regular sintered disks	
	110806-37	110806-38	110806-39	110702-12	110706-25
Mass (g)	1.072	1.073	1.069	1.077	1.068
Packing	**91.0%**	**91.7%**	**91.5%**	**91.7%**	**91.6%**
dissolution time, min	1.33	1.43	1.35	**25**	**11**
% dissolved	100.0%	100.0%	100.0%	**77.8%**	100.0%
remainings, g	N/A	N/A	N/A	0.2393	N/A
Diss. rate, g/min	**0.804**	**0.749**	**0.792**	**0.034**	**0.097**

Conversion to alkaline solution

$$2[Mo_2O_3(O_2)_4(H_2O)_2]^{2-} = 4MoO_3 \cdot H_2O + 5O_2 \ (H_2O_2 \text{ decomposition})$$

$$2[Mo_2O_3(O_2)_4(H_2O)_2]^{2-} + 8KOH = 4K_2MoO_4(H_2O)_2 + 5O_2$$

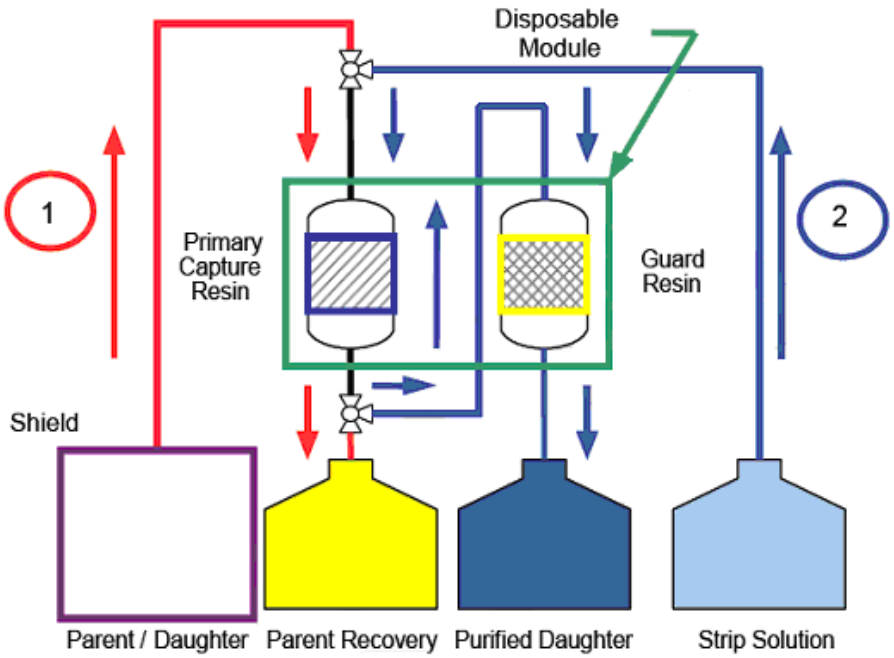
To convert 1g of Mo to K_2MoO_4 we need ~1.17g of KOH (~87% KOH – 1.34g KOH)
~6.7g of KOH/20mL for 6M KOH



	Solubility of Mo, g/100mL	
Concentration	NaOH	KOH
4M	15.7	28.2
5M	12.9	26.5
6M	-	24.9

~80% of max. solubility – 20g/100mL

Mo/Tc separation – NorthStar generator (pre-dates TechneGen)

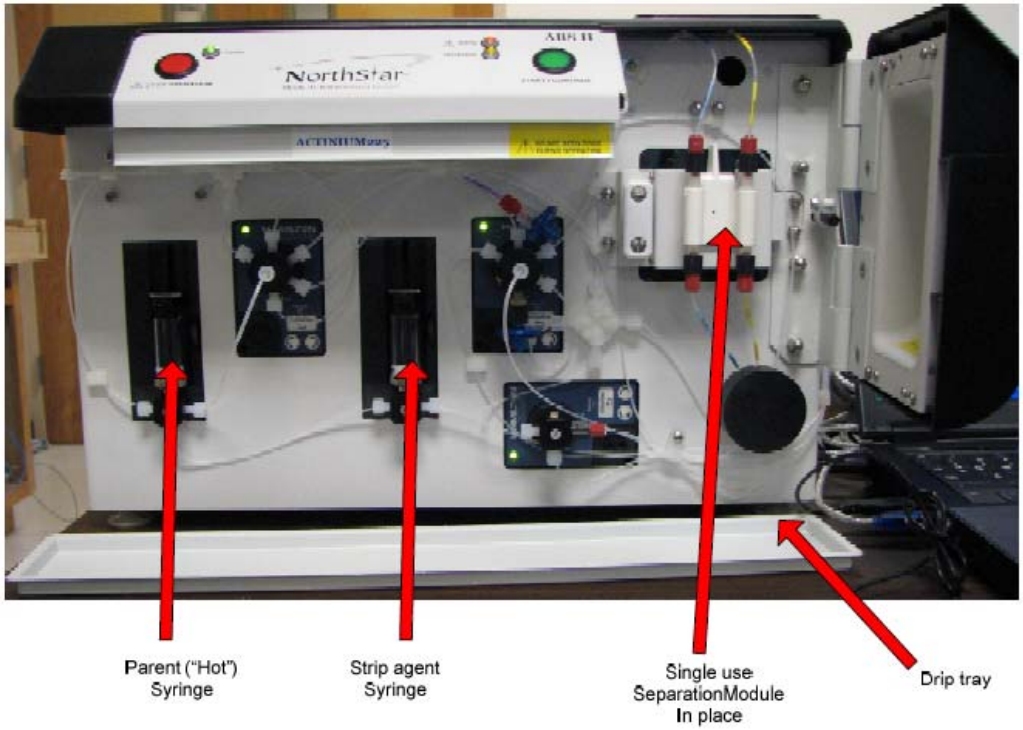


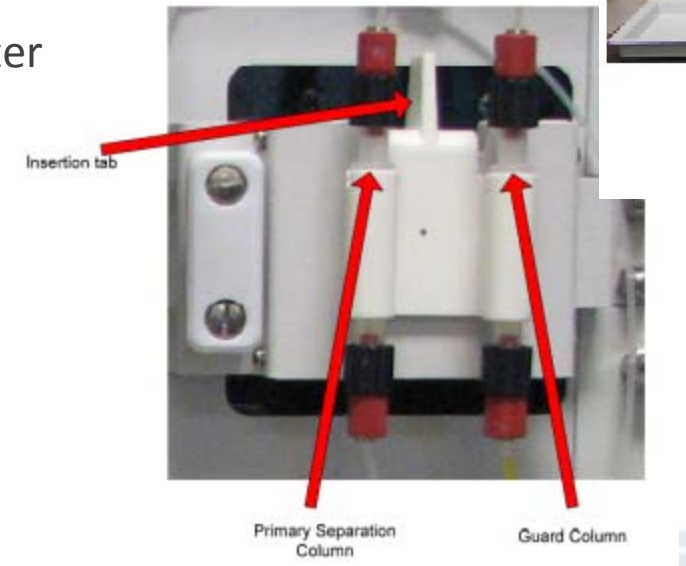
- Tc sorbs on ABEC column in alkaline solution
- Mo recovered in parent recovery bottle
- ABEC column rinsed with (Na/K)OH and buffer solution
- Tc stripped by normal saline solution
- Guard Column (Alumina) selective for Mo
- Tc recovered in product vial

Mo/Tc separation – NorthStar generator

Monitoring of Mo and Tc throughout the generator

- Tc Product vial
  - Mo, Tc activity, pH, Al content
- Mo, Tc activity monitoring
  - ABEC Column
  - Guard Column
  - Waste solution
  - Filter

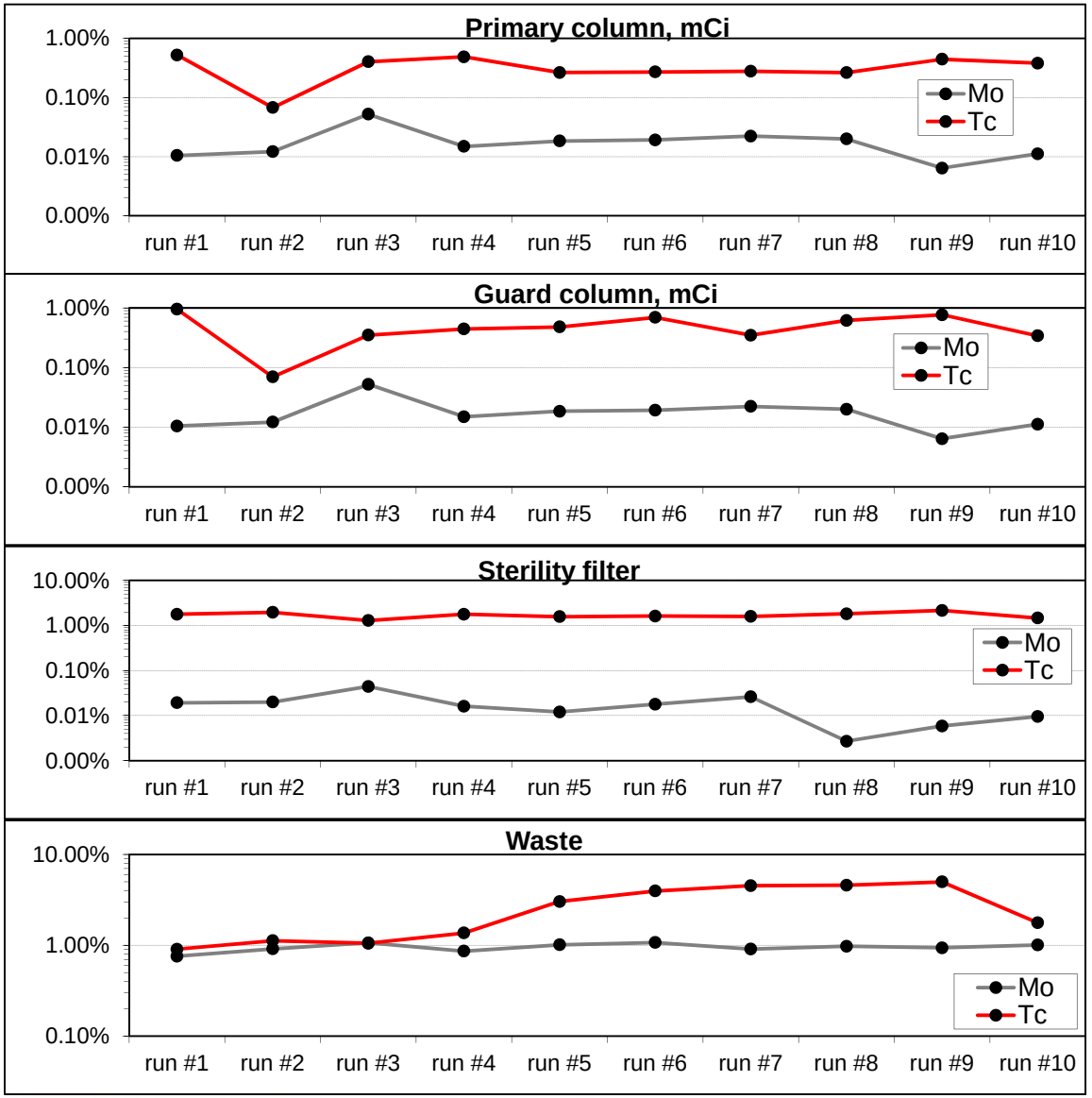




Mo/Tc separation – NorthStar generator

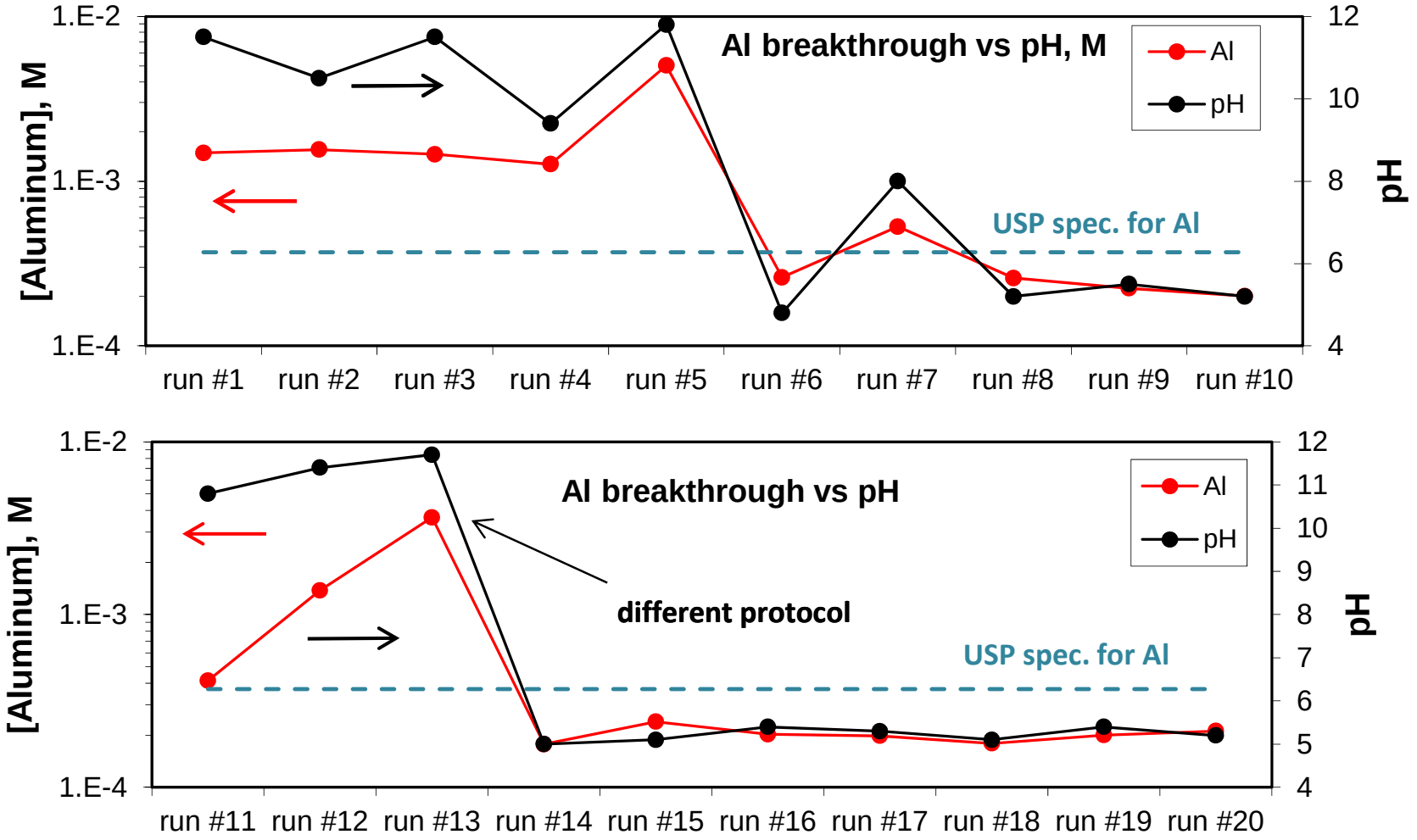
3 tests of ARS-II performed

- **First test – 8 runs**
  - Tc recovery (88-95%)
- **Second test – 8 runs**
  - Tc recovery (88-95%)
  - Mo and Tc activity
  - pH=11-13
- **Third test – 20 runs**
  - Average ~95% recovery
  - Mo and Tc activity
  - Mo and Al breakthrough
  - pH control
  - Mo and Al meet USP spec. if pH<5.5



Mo/Tc separation – Al content in the Tc product

USP specification for Al content in Tc product (<10 ppm, or <3.71 × 10^{-4} M aluminum).



Future plans

- Radiolytic stability of ABEC column (up to 54kGy previously tested)
- Thermal and production experiments
- Evaluate reliability of the He-cooling system
- Production of several curies of Mo-99 (~20Ci)
- Post irradiation processing of Mo disks
- Testing TechneGen Generator System

Acknowledgement

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. Argonne National Laboratory is operated for the U.S. Department of Energy by UChicago Argonne, LLC.