Production of radioisotopes using secondary neutrons at the Brookhaven Linac Isotope Producer

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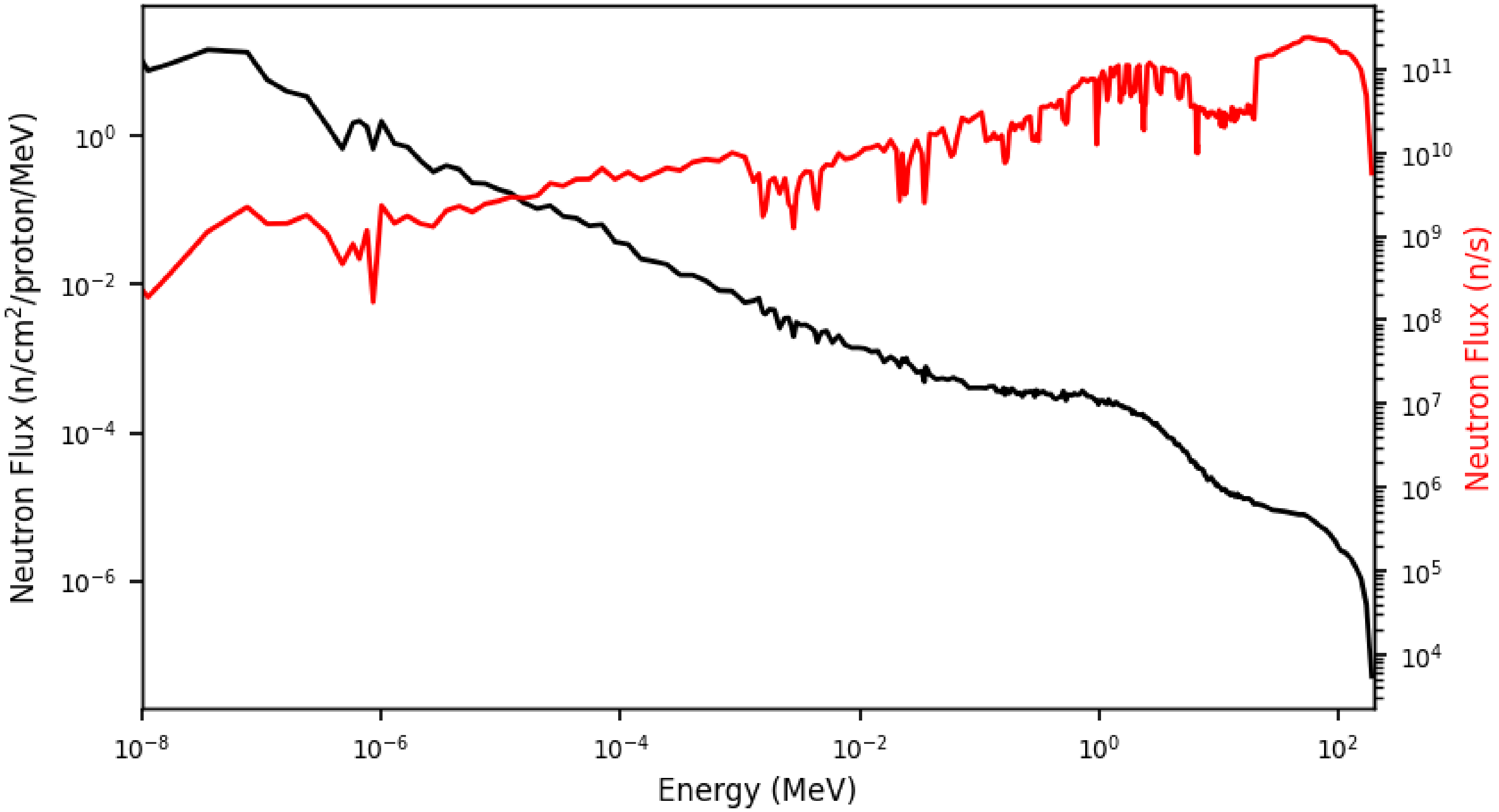
Neutron Production of Radioisotopes

- Alternative production routes for radionuclides compared to charged-particle reactions
- No Coulomb barrier to overcome (no charge)
- Neutrons come free from proton-induced reactions on a production target at BLIP
  - Constant flux from proton-irradiated target stack upstream
  - Large flux of fast neutrons >14 MeV unavailable at reactors

Neutron Spectrum from FLUKA



Brookhaven National Laboratory

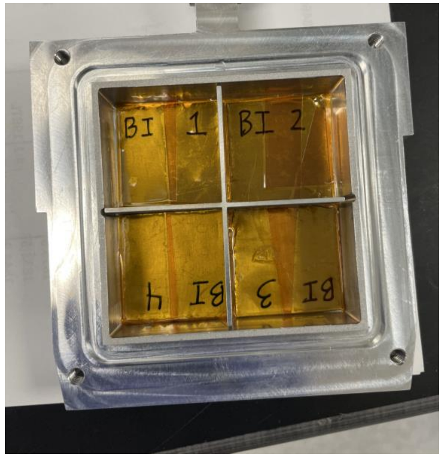
Irradiation Campaigns

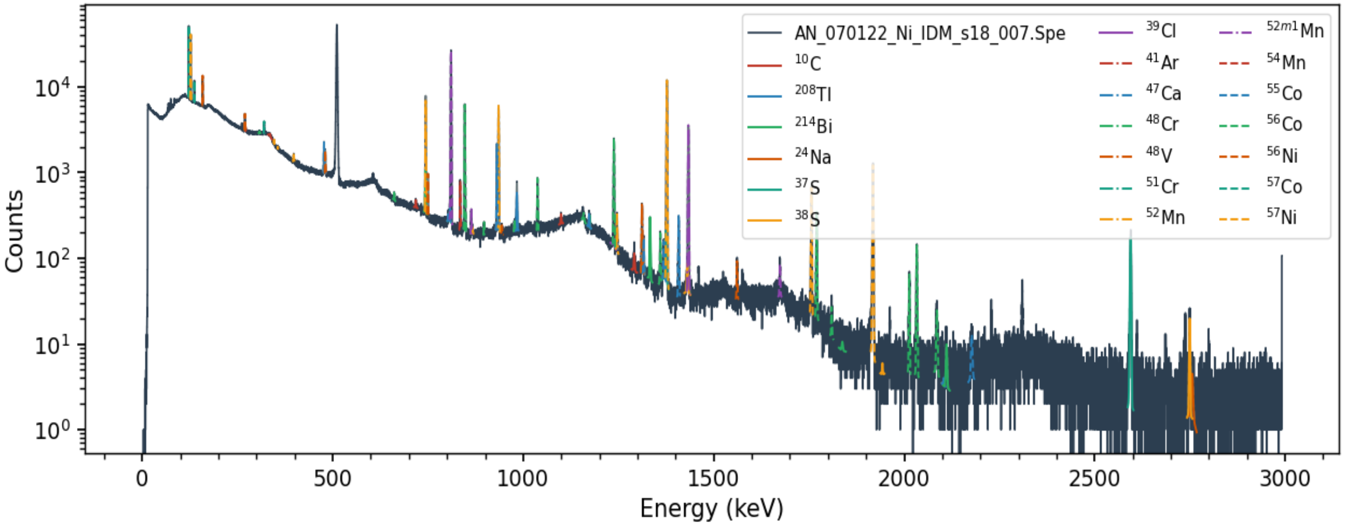
Date	Energy (MeV)	Current (μA·h)	Foils	Goal
June 2021	117	2,700	Ti, Co, Bi	Evaluation of producing select radionuclides in the n-slot
February 2022	200	300	Ni, Bi	Determining inhomogeneity in quadrants of the n-slot
June 2022	200	75	Al, Co, Ni, Zn, Y, Au, Bi	Calculation of neutron spectrum using radionuclides produced over full energy range

<u>Experimental Procedure</u>

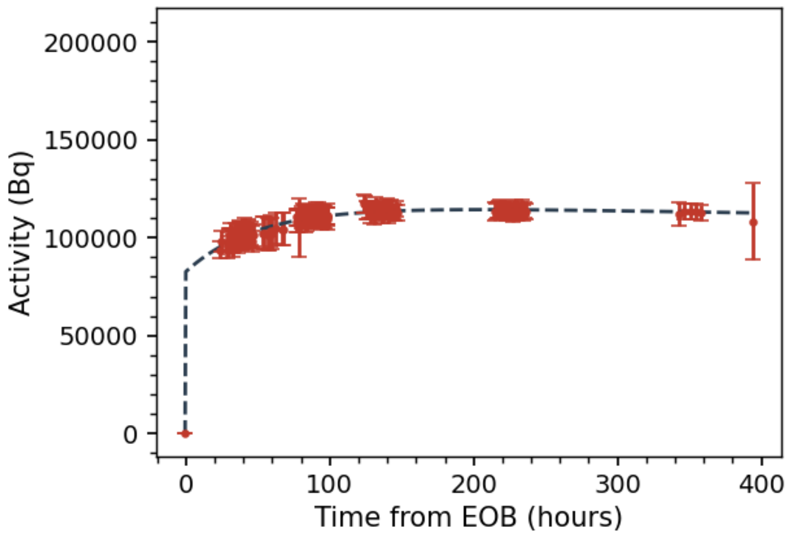
1. Irradiation of foils at BLIP using neutrons behind a target array
2. Measurement of gamma rays from radionuclide decay
3. Data analysis for determination of activities/production rates

Irradiation, Measurement, & Analysis





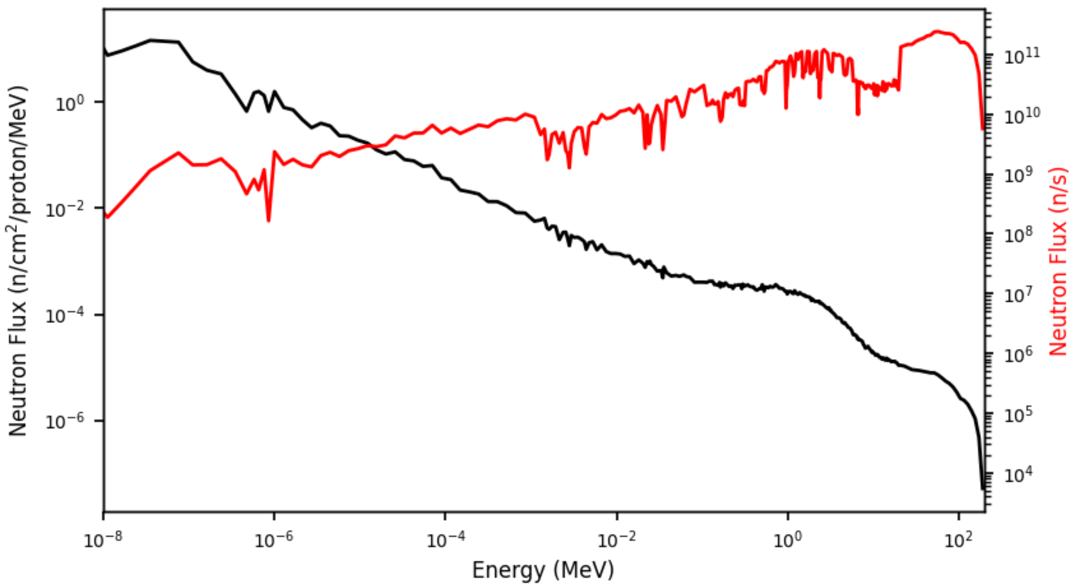
57gCo



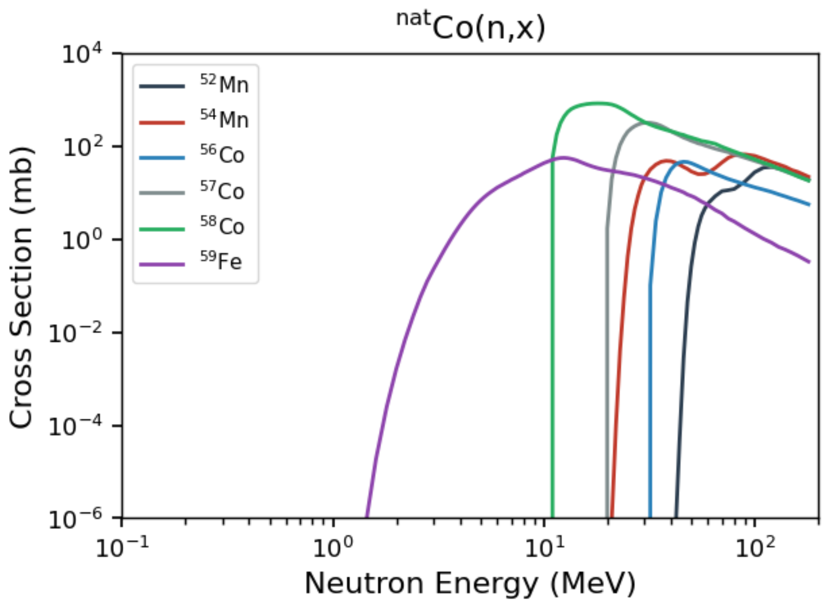
Neutron Spectrum Calculation

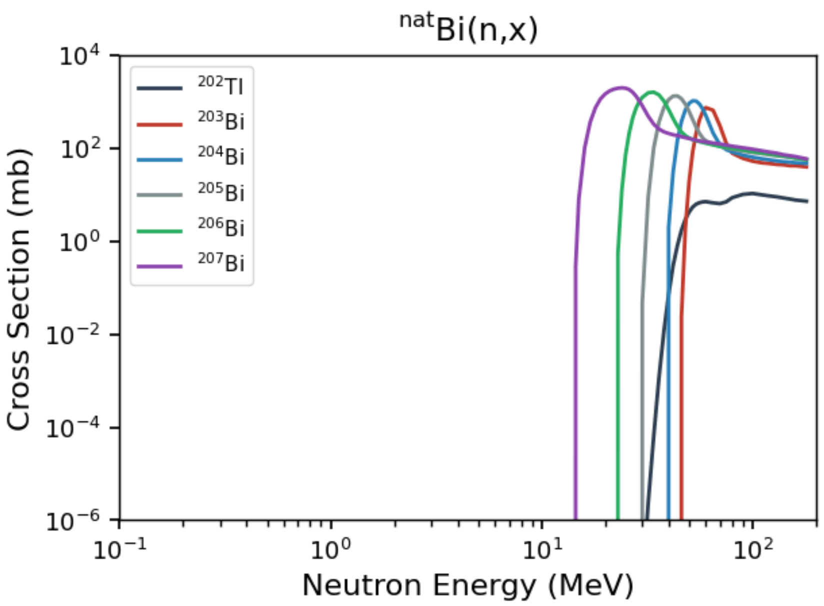
$$R = \int N \times \sigma(E) \times \Phi(E)$$

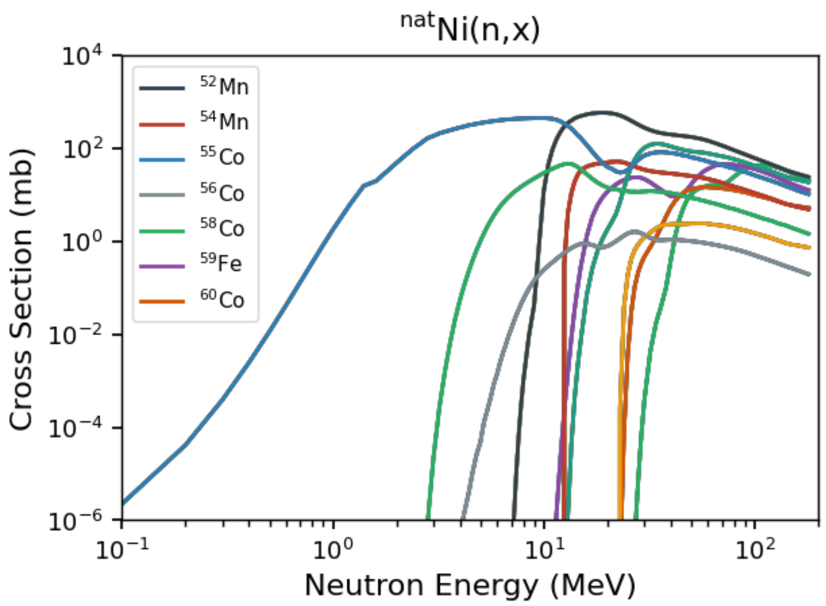
- Discrete energy points for cross section
- Cross sections from TENDL
- Monte Carlo variation of Φ(E)



Cross Sections for Monitor Isotopes



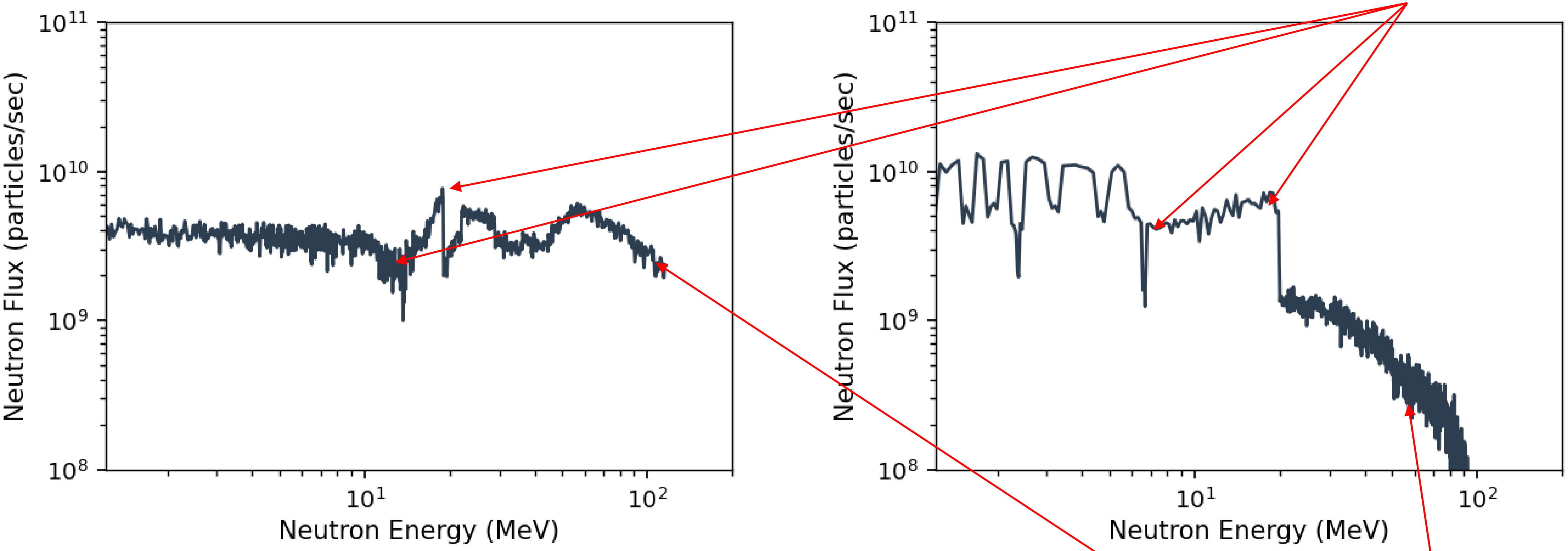




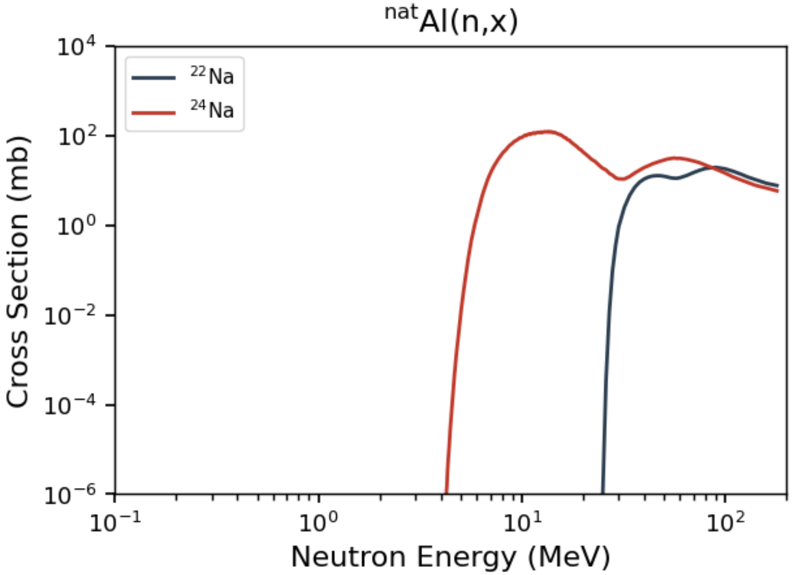
Results of MC Calculation

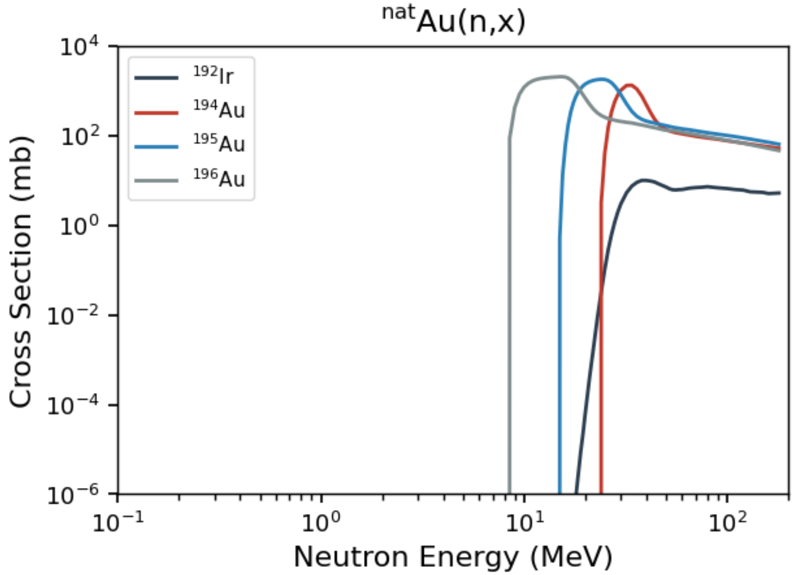
(June 2021)

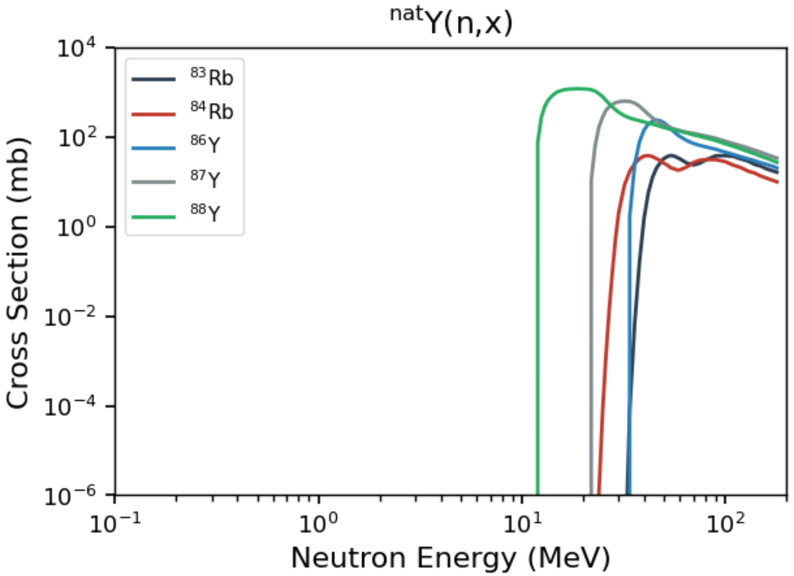
Similar features that correspond to nuclear phenomena

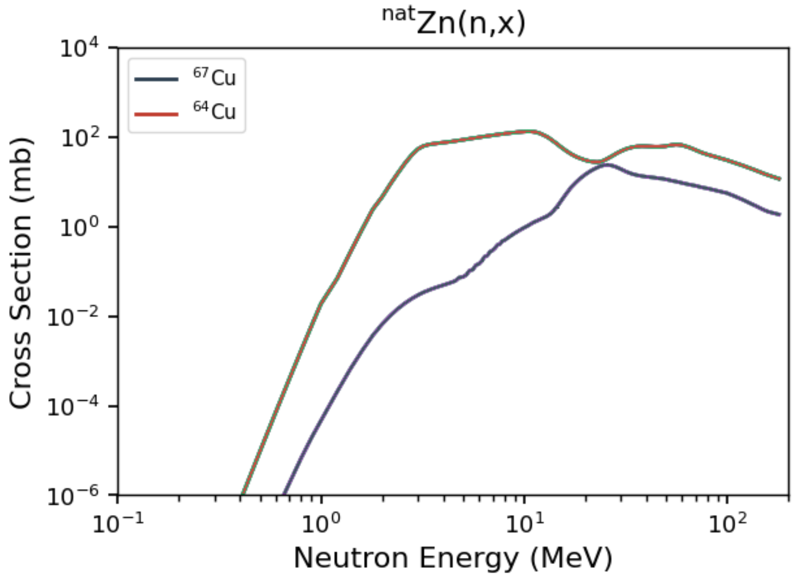


Other features may reflect low statistics or reliance on one or two cross sections


natAl(n,x)
22Na
24Na
Cross Section (mb)
Neutron Energy (MeV)


natAu(n,x)
192Ir
194Au
195Au
196Au
Cross Section (mb)
Neutron Energy (MeV)

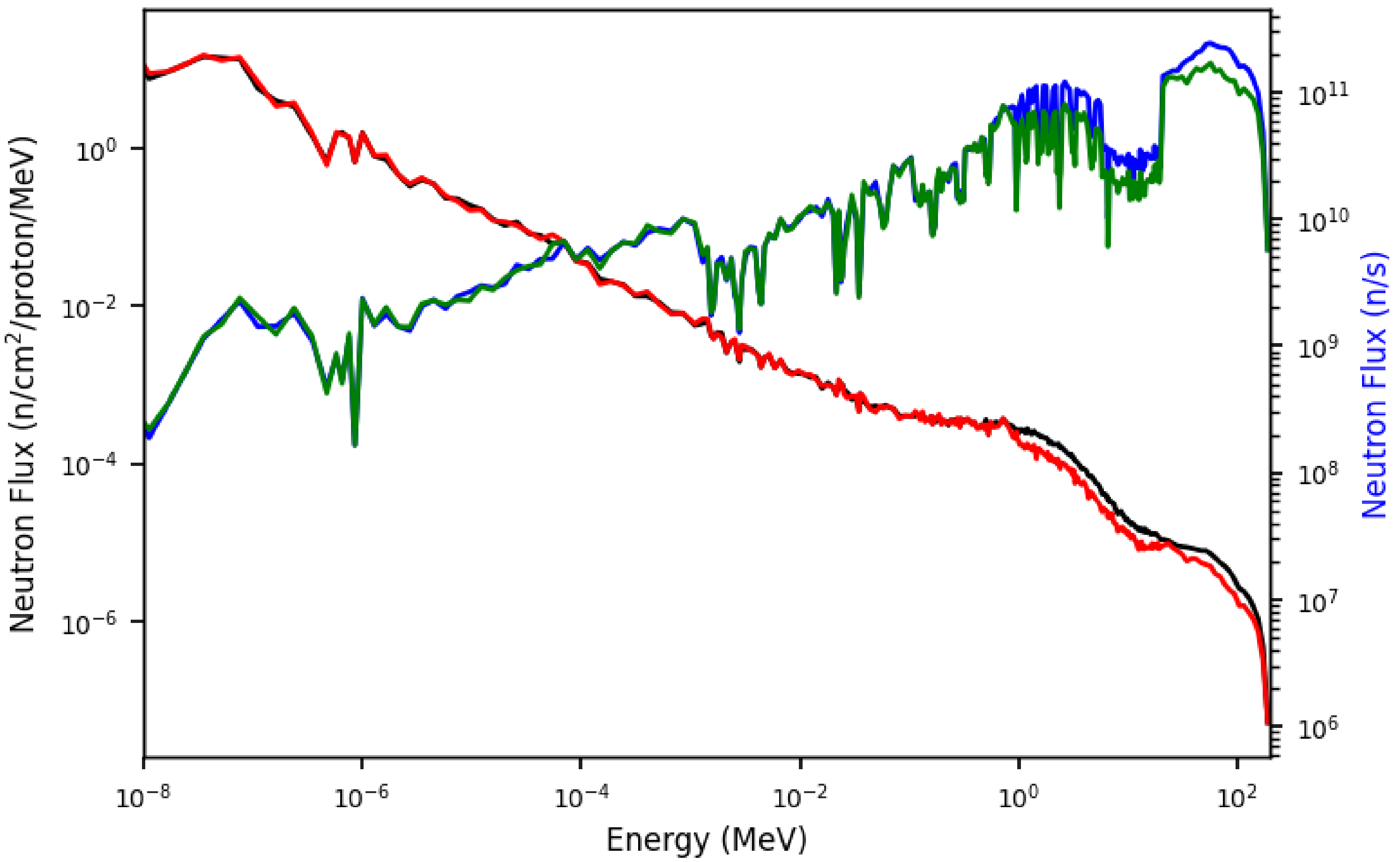

natY(n,x)
83Rb
84Rb
86Y
87Y
88Y
Cross Section (mb)
Neutron Energy (MeV)


natZn(n,x)
67Cu
64Cu
Cross Section (mb)
Neutron Energy (MeV)

Modified Neutron Spectrum

FLUKA Flux (n/cm²/MeV/proton)

Modified Flux (n/cm²/MeV/proton)



FLUKA Flux (n/s)

Modified Flux (n/s)

Radionuclide Production

Date	Energy (MeV)	Current (μA)	Irradiation Time (h)	Activity at EOB (μCi)		Saturation Rate (μCi/μA/g)	
				^{47}Sc	^{59}Fe	^{47}Sc*	^{59}Fe
June 2021	117	116	23	1.39×10^3	44.81	94.77	18.62
June 2022	200	150	0.5	not in n-slot	0.77	not in n-slot	28.76

^{47}Sc produced from Ti foil, ^{59}Fe produced from Co foil
*co-production of ^{48}Sc is a challenge to radionuclidic purity

Summary

- Neutrons provide a way of producing additional radioisotopes that may not be accessible through other means, and are already being generated at BLIP with higher energies than those available at reactor facilities

- The initial tests for mapping the neutron spectrum from BLIP has gone successfully, and the irradiation this past June will help solidify the neutron flux behind BLIP for newer target arrays

- Production of medical isotopes such as ^{47}Sc, ^{59}Fe has been demonstrated, and future medical isotopes will be explored in this manner using natural or enriched targets

Acknowledgments

- Experimental/Safety support
  - MIRP/BLIP Operators
    - Lisa Muench
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    - David O’Rourke
  - Radiation Control Division
    - Pat Sullivan
    - Vicky Litton
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  - US Department of Energy Isotope Program
- BNL is managed by Brookhaven Science Associates operated by Battelle Memorial Institute (Battelle) and Stony Brook University for US DOE Office of Science

Backup Slides

BLIP


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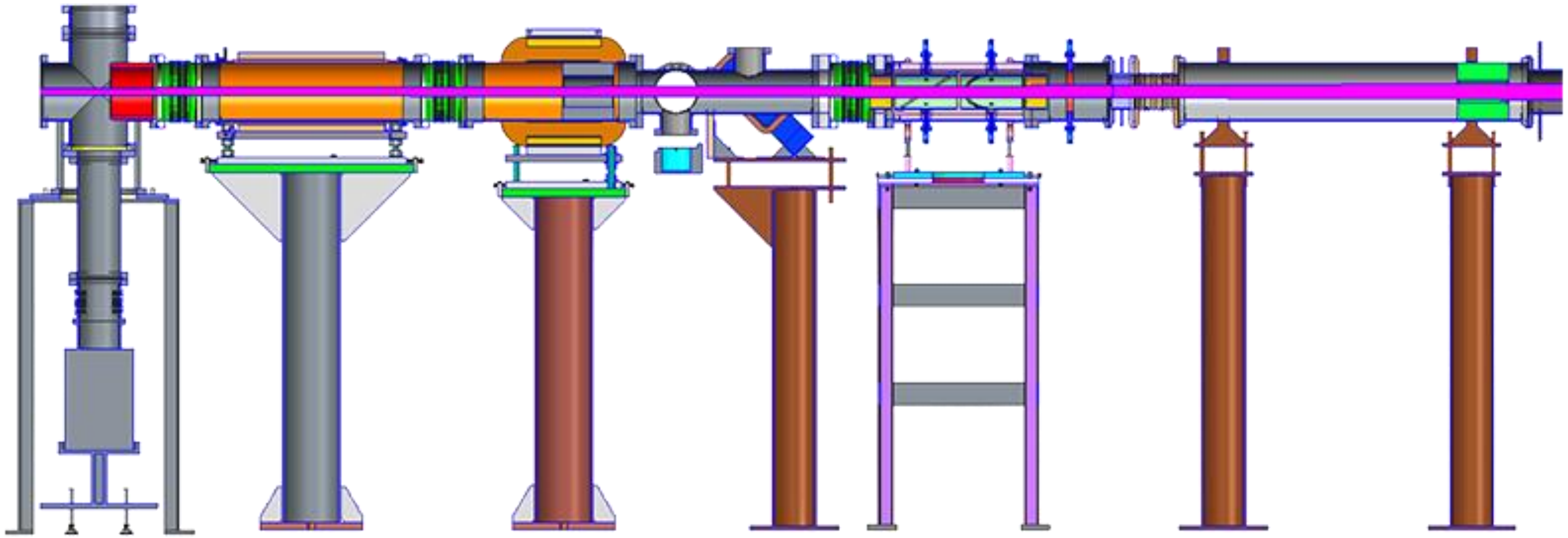
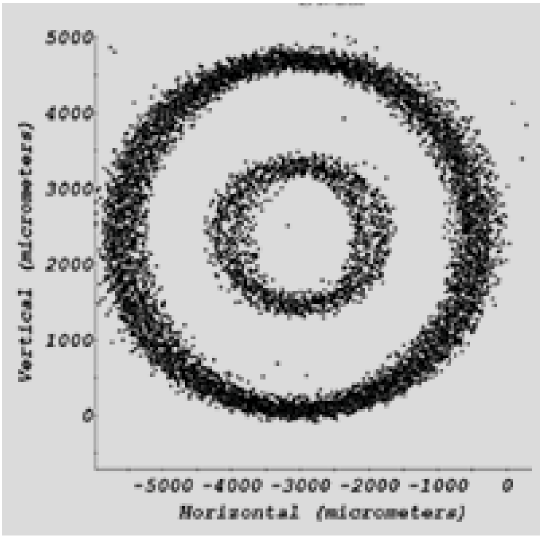



5000
4000
3000
2000
1000
0
Vertical (micrometers)
-5000 -4000 -3000 -2000 -1000 0
Horizontal (micrometers)

Irradiation of Foils – June 2021

- Bi, Co, Ti foils chosen for irradiation
  - 25 mm x 25 mm, 0.25 mm thick
- 23 hour proton irradiation at 116 µA, 117 MeV, rastered beam
  - Time maximizes production of isotopes to 200 mCi control limit which includes short-lived isotopes (minute half-lives)
- Foils counted at BLIP after cooling period

Post-irradiation

- Foils were too hot to be counted right away (0.25 – 1.3 R/h on contact)
  - Each foil cooled until the 150 mrem/h limit for rad work was reached

- Bi foil was so hot that unfortunately the $^{203,204}Bi$ (~11.5 h half-lives) activities could not be measured before 150 mR/h was reached

June 2021

- 25 mm x 25 mm, 0.25 mm thick foils
- 23 hour proton irradiation
  - 116 µA, 117 MeV, rastered
- Foils counted at BLIP after cooling period

Isotope	$t_{1/2}$ (days)	Activity
Bi Foil		
^{205}Bi	15.31	0.29 mCi
^{206}Bi	6.24	1.03 mCi
^{207}Bi	1.15×10^{4}	1.27 µCi
Co Foil		
^{56}Co	77.24	14.49 µCi
^{57}Co	271.74	32.45 µCi
^{58}Co	70.86	0.38 mCi
^{60}Co	1925.28	0.33 mCi
^{59}Fe	44.495	44.81 µCi
Ti Foil		
^{46}Sc	83.79	46.98 µCi
^{47}Sc	3.35	1.39 mCi
^{48}Sc	1.82	0.69 mCi

Produced radioisotopes (EOB)

Isotope	$t_{1/2}$ (days)	Activity
Bi Foil		
^{205}Bi	15.31	0.29 ± 0.03 mCi
^{206}Bi	6.24	1.03 ± 0.08 mCi
^{207}Bi	1.15×10^{4}	1.27 ± 0.05 µCi
Co Foil		
^{56}Co	77.24	14.49 ± 0.72 µCi
^{57}Co	271.74	32.45 ± 0.83 µCi
^{58}Co	70.86	0.38 ± 0.01 mCi
^{60}Co	1925.28	0.33 ± 0.01 mCi
^{59}Fe	44.495	44.81 ± 18.13 µCi
Ti Foil		
^{46}Sc	83.79	46.98 ± 1.61 µCi
^{47}Sc	3.35	1.39 ± 0.10 mCi
^{48}Sc	1.82	0.69 ± 0.03 mCi

Produced radioisotopes vs FLUKA

Bi Foil

Isotope	$t_{1/2}$ (days)	Activity	Predicted / Measured
^{205}Bi	15.31	0.29 ± 0.03 mCi	0.97
^{206}Bi	6.24	1.03 ± 0.08 mCi	0.81
^{207}Bi	1.15×10^4	1.27 ± 0.05 µCi	0.8

Ti Foil

Isotope	$t_{1/2}$ (days)	Activity	Predicted / Measured
^{46}Sc	83.79	46.98 ± 1.61 µCi	0.53
^{47}Sc	3.35	1.39 ± 0.10 mCi	0.46
^{48}Sc	1.82	0.69 ± 0.03 mCi	0.21

Co Foil

Isotope	$t_{1/2}$ (days)	Activity	Predicted / Measured
^{56}Co	77.24	14.49 ± 0.72 µCi	1.46
^{57}Co	271.74	32.45 ± 0.83 µCi	0.74
^{58}Co	70.86	0.38 ± 0.01 mCi	0.83
^{60}Co	1925.28	0.33 ± 0.01 mCi	0.07
^{59}Fe	44.495	44.81 ± 18.13 µCi	0.93

Irradiation – February 2022

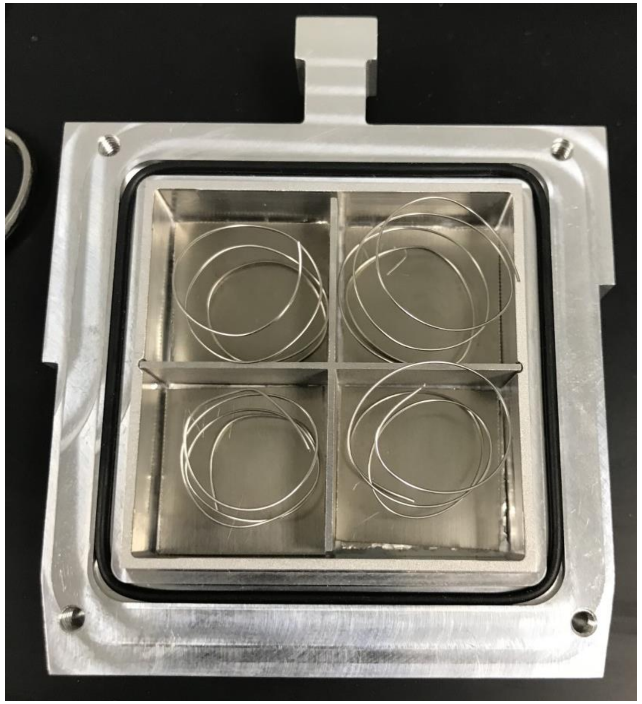
Irradiation of Bi, Ni foils in all four quadrants of n slot box to help identify if there are appreciable differences in flux

- Contributable to slight beam misalignment, or beam propagation from start of the stack to the end

150 µA, 200 MeV protons for 2 hours of irradiation behind a thorium target array – neutron spectrum will better reflect real BLIP environment

New monitor isotopes from Ni foils cover a wider energy range than Co, including a zero-threshold reaction, and have larger cross sections

Conducted late February, data collecting and will be analyzed in the upcoming weeks, but ^{203}Bi and ^{204}Bi have been captured



Irradiation – June 2022

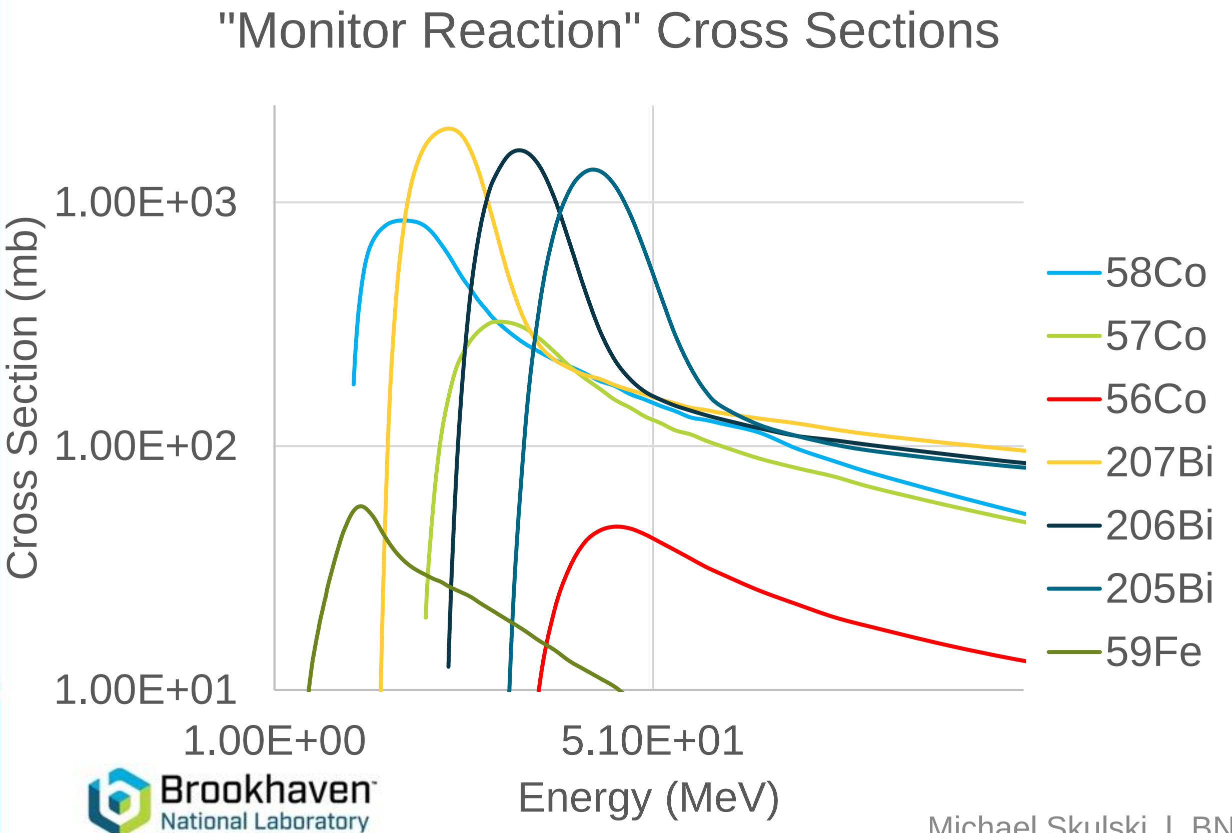
- 7 elements (natural abundance)

- 3 planes of foils
    1. 4 Bi for tracking flux distribution
    2. Al, Ni, Zn
    3. Co, Y, Au

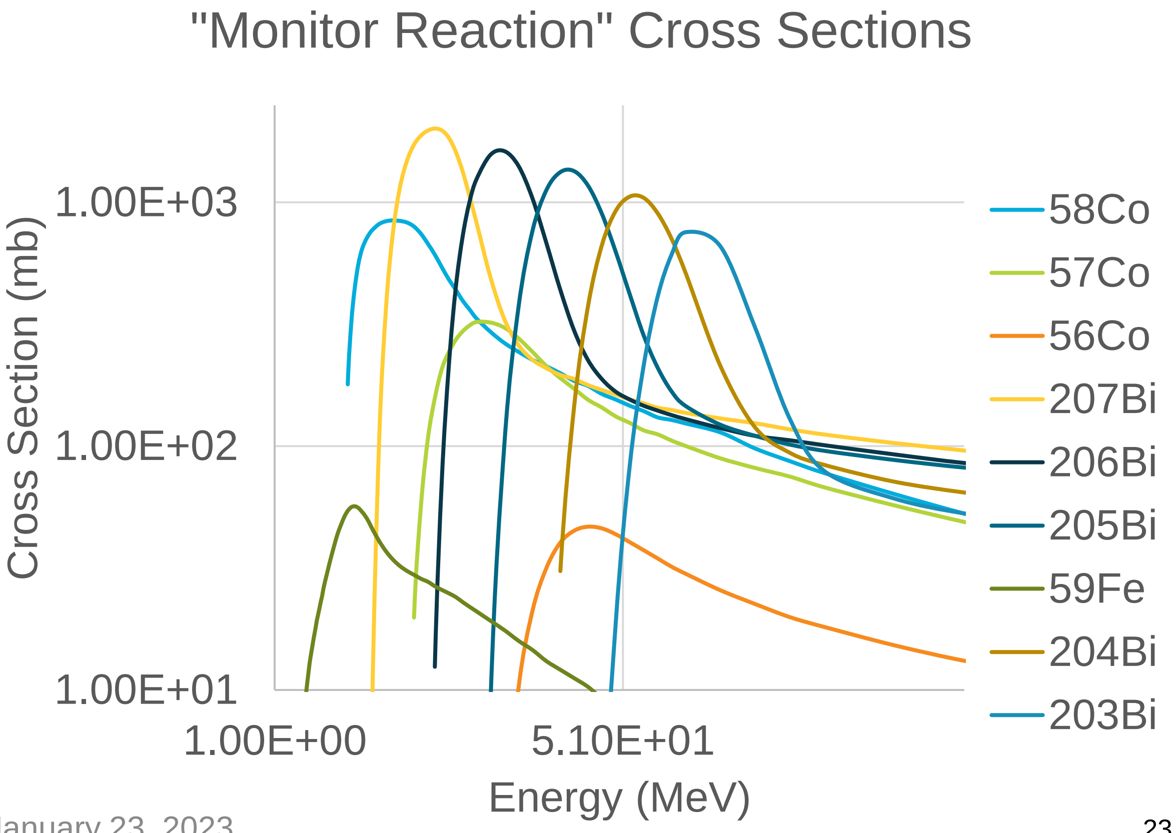
- Proton irradiation of thorium target array
    - 200 MeV
    - 150 µA
    - 30 minutes

Monitor Reaction Cross Sections

Currently Possible because of Bi cooling period



Possible with Shorter/Lower Current Irradiation



New Monitor Products

Radionuclide	$t_{1/2}$ (days)	Activity
Zn Foil		
^{67}Cu	2.6	15.57 ± 0.05 µCi
Y Foil		
^{86}Y	0.6	131.82 ± 0.36 µCi
^{88}Y	106.6	1.99 ± 0.01 µCi
Au Foil		
^{194}Au	1.6	201.05 ± 11.01 µCi
^{196}Au	6.2	69.87 ± 0.90 µCi