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

Michael Skulski, Dmitri G. Medvedev, Cathy S. Cutler

RaDIATE Collaboration Meeting – June 27, 2023

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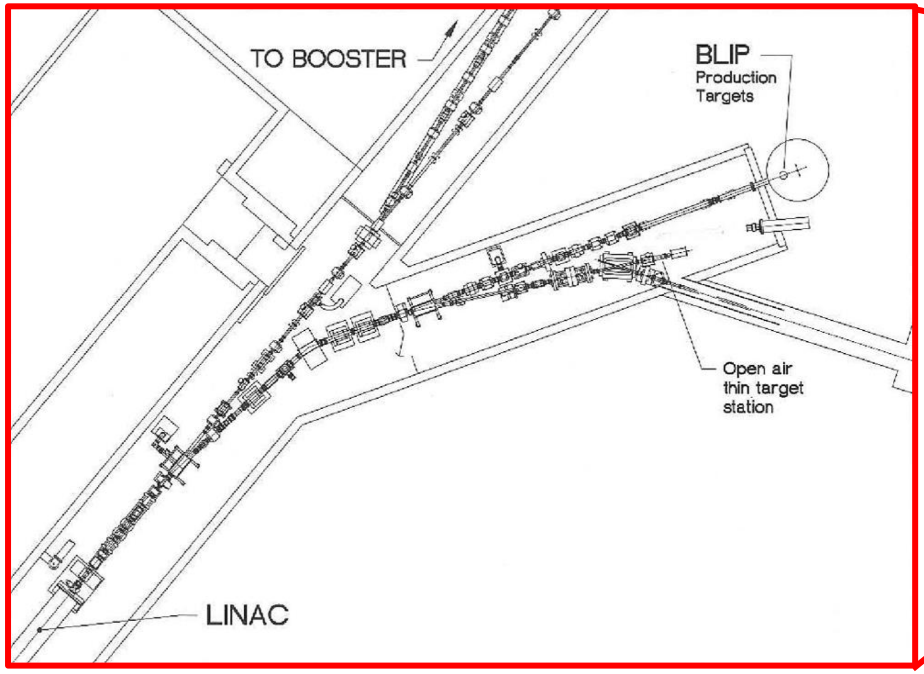
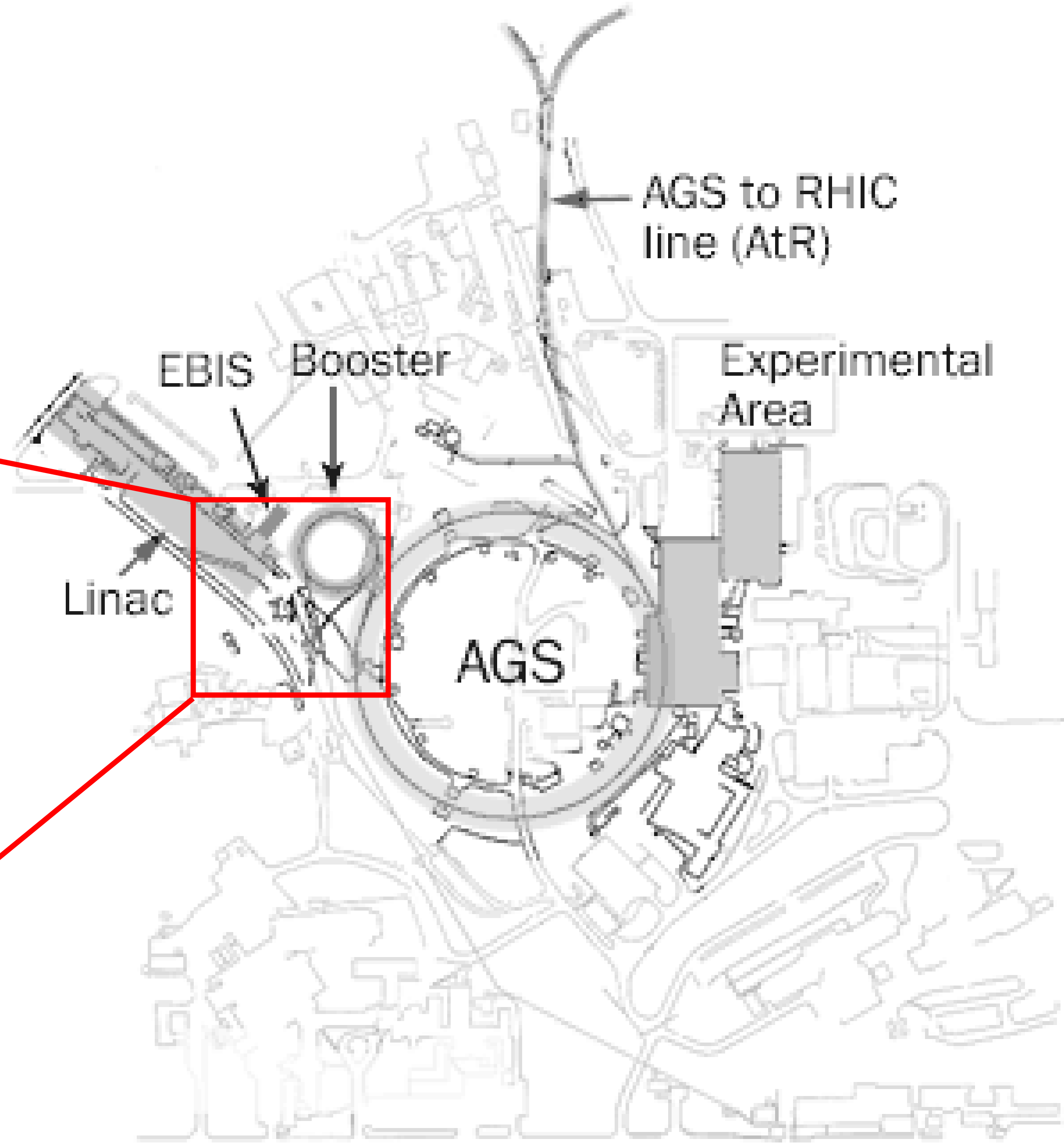
200 MeV Linac





BLIP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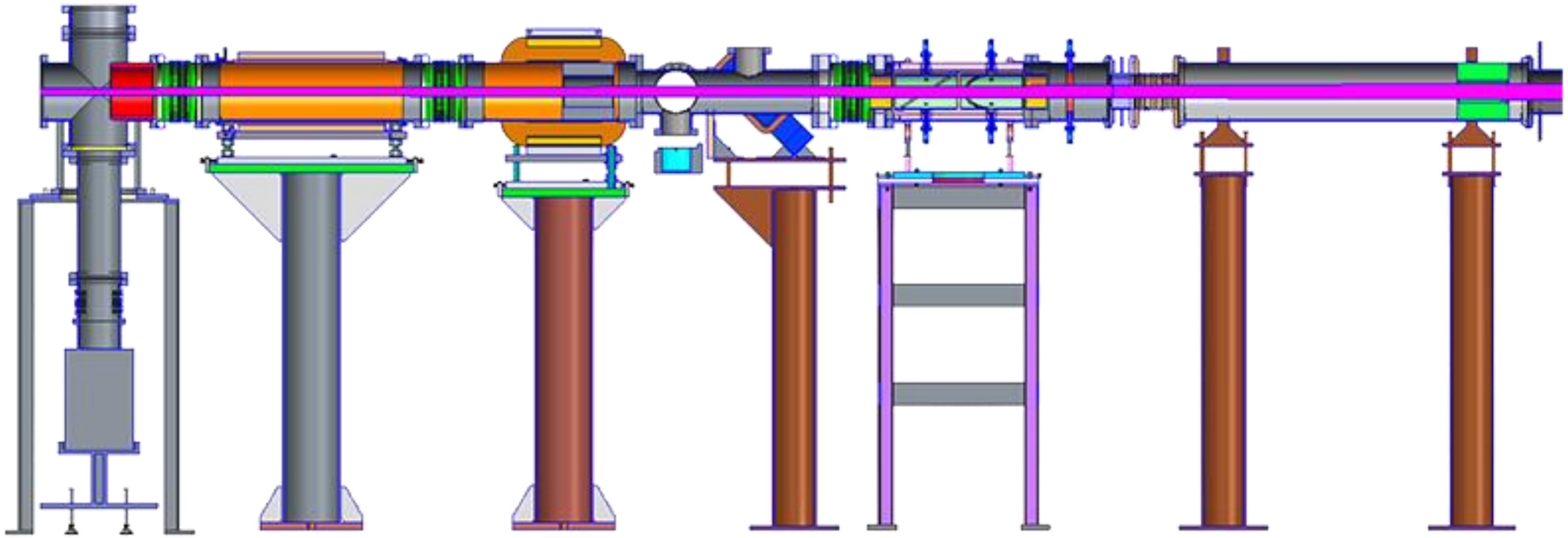
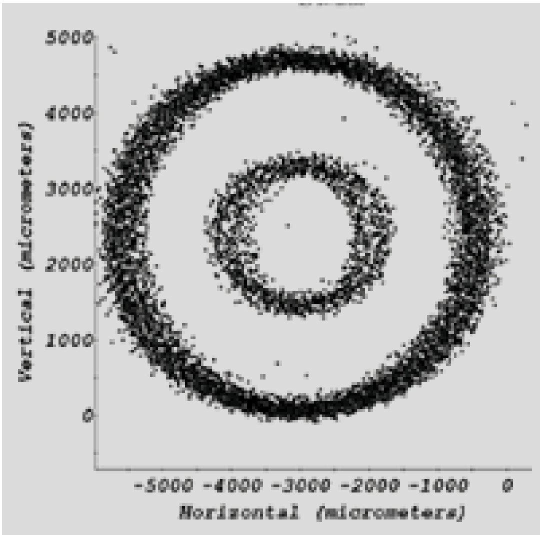



5000
4000
3000
2000
1000
0
Vertical (micrometers)
-5000 -4000 -3000 -2000 -1000 0
Horizontal (micrometers)

Neutron Production of Radioisotopes

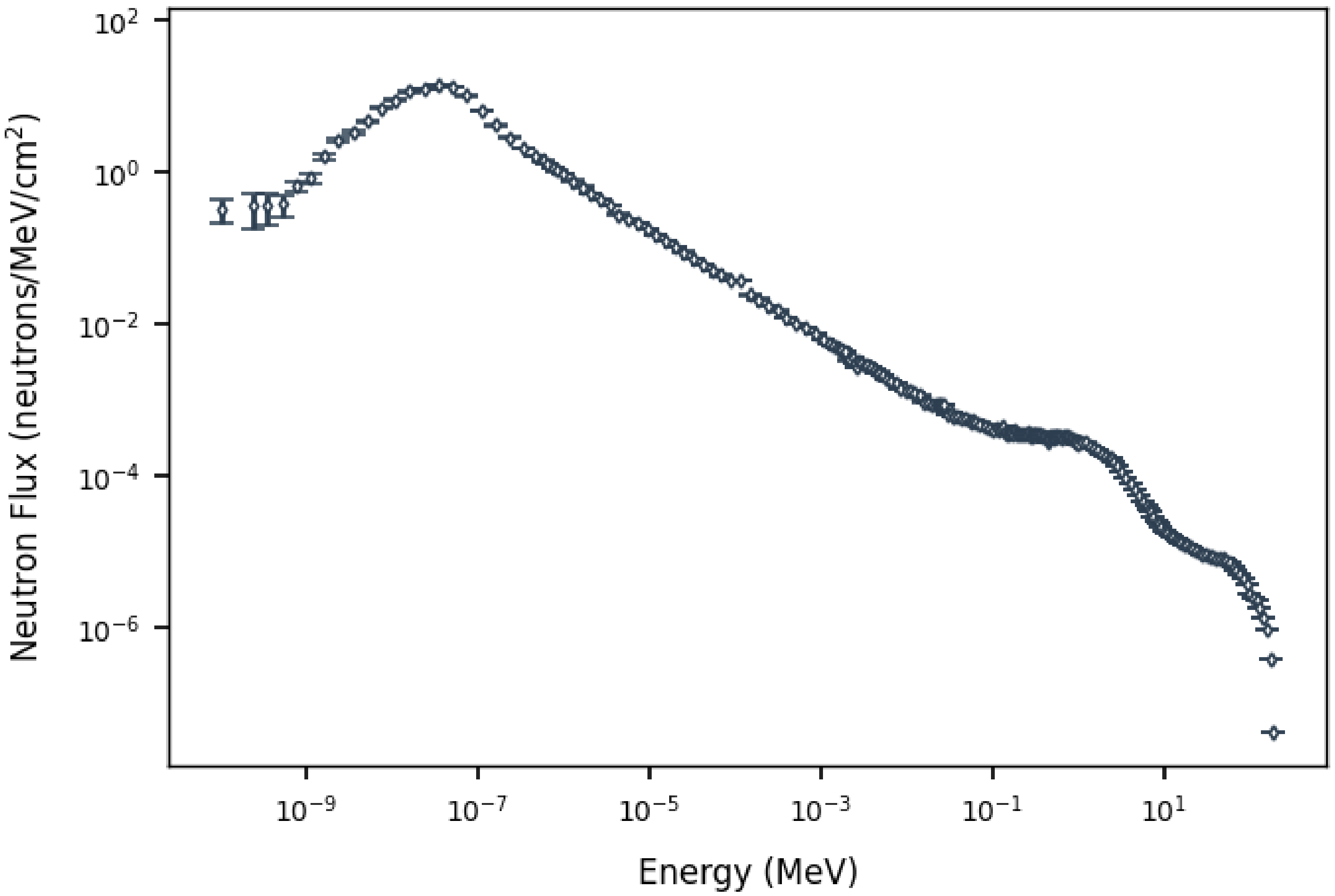
Why Neutrons?

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

Challenges?

- Energy is a spectrum, not a (quasi-) monoenergetic beam
- Emitted from targets in an angular distribution

FLUKA Simulation



Strategy

Work at LANL (Engle/Mosby)* as a guide to initial irradiations

1. Irradiation of target stack

2. Quantification of activities (flux monitors, medical isotopes)

3. Calculation of neutron flux using nuclear data

*NIM A, Volume 754, 1 August 2014, Pages 71-82
*NIM B, Volume 381, 15 August 2016, Pages 29-33

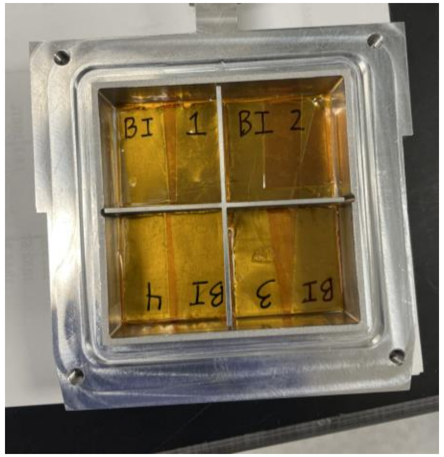
Irradiation Campaigns

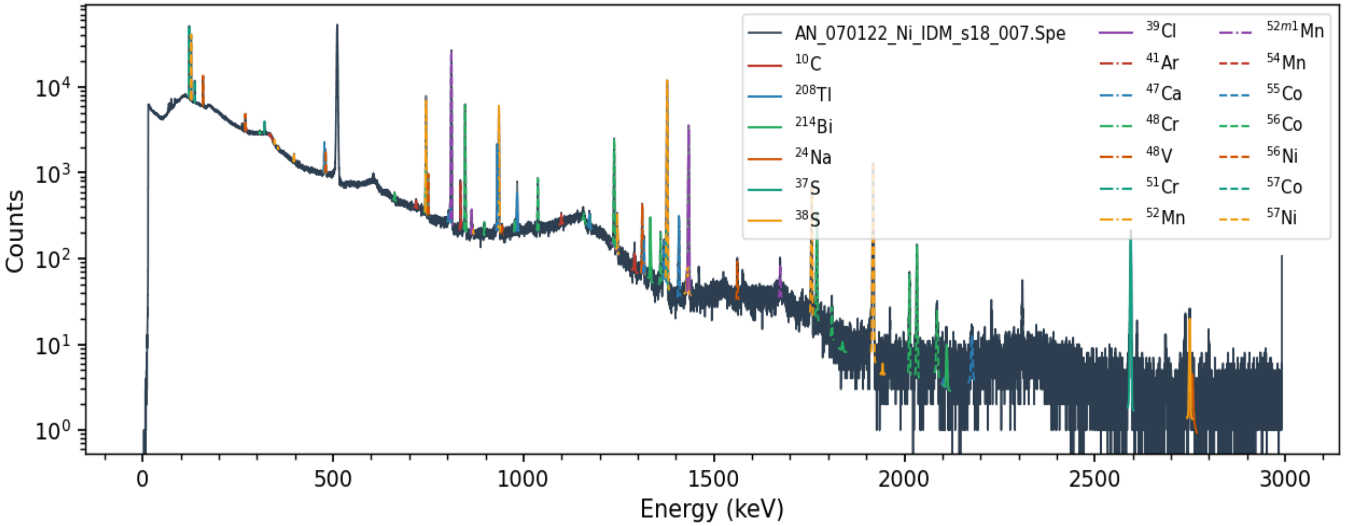
Date	Energy (MeV)	Current (μA·h)	Foils	Goal
June 2021	117	2,700	Ti, Co, Bi	Evaluation of producing select radionuclides in the n-slot
February 2022	200	300	Ni, Bi	Determining inhomogeneity in quadrants of the n-slot
June 2022	200	75	Al, Co, Ni, Zn, Y, Au, Bi	Calculation of neutron spectrum using radionuclides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

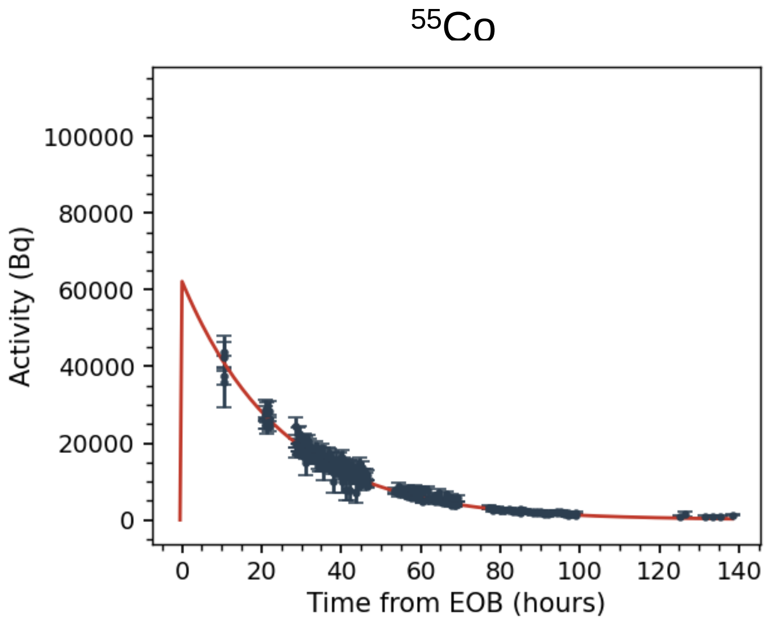
Experimental Procedure

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







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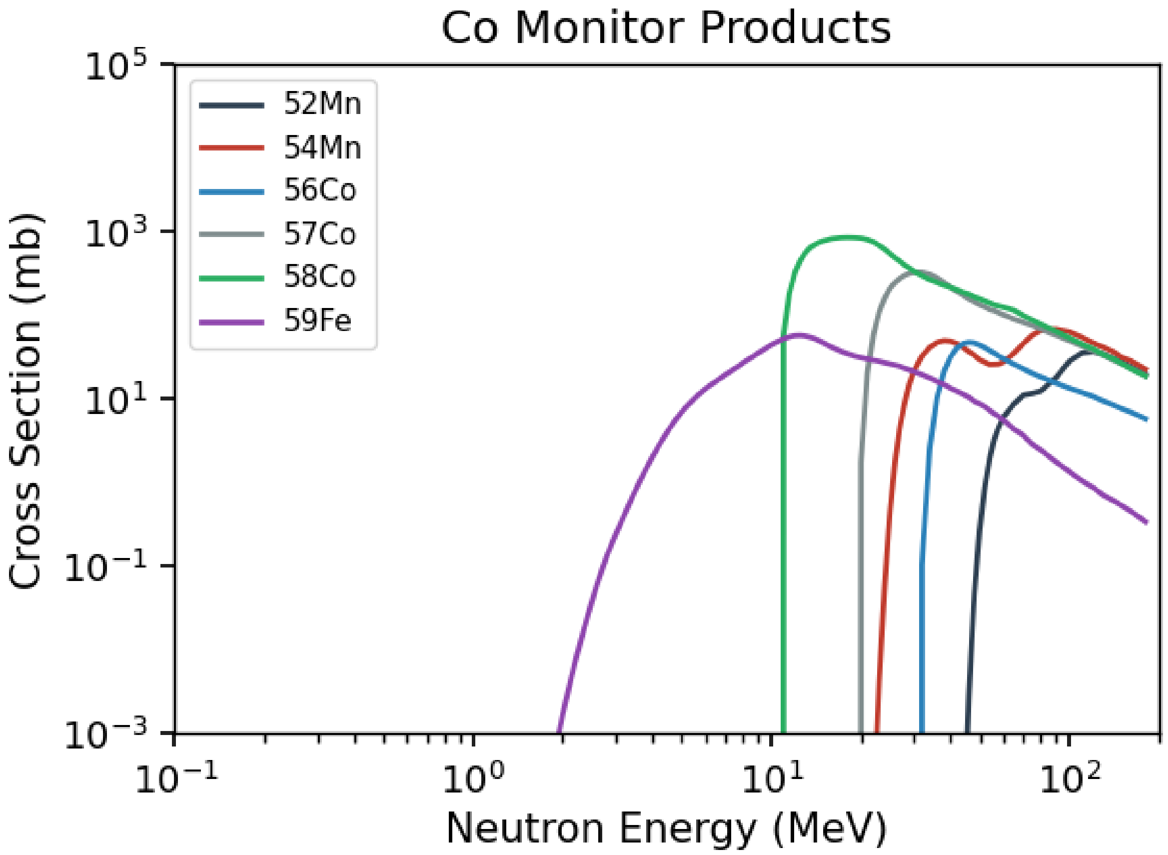
Irradiation of Foils – June 2021

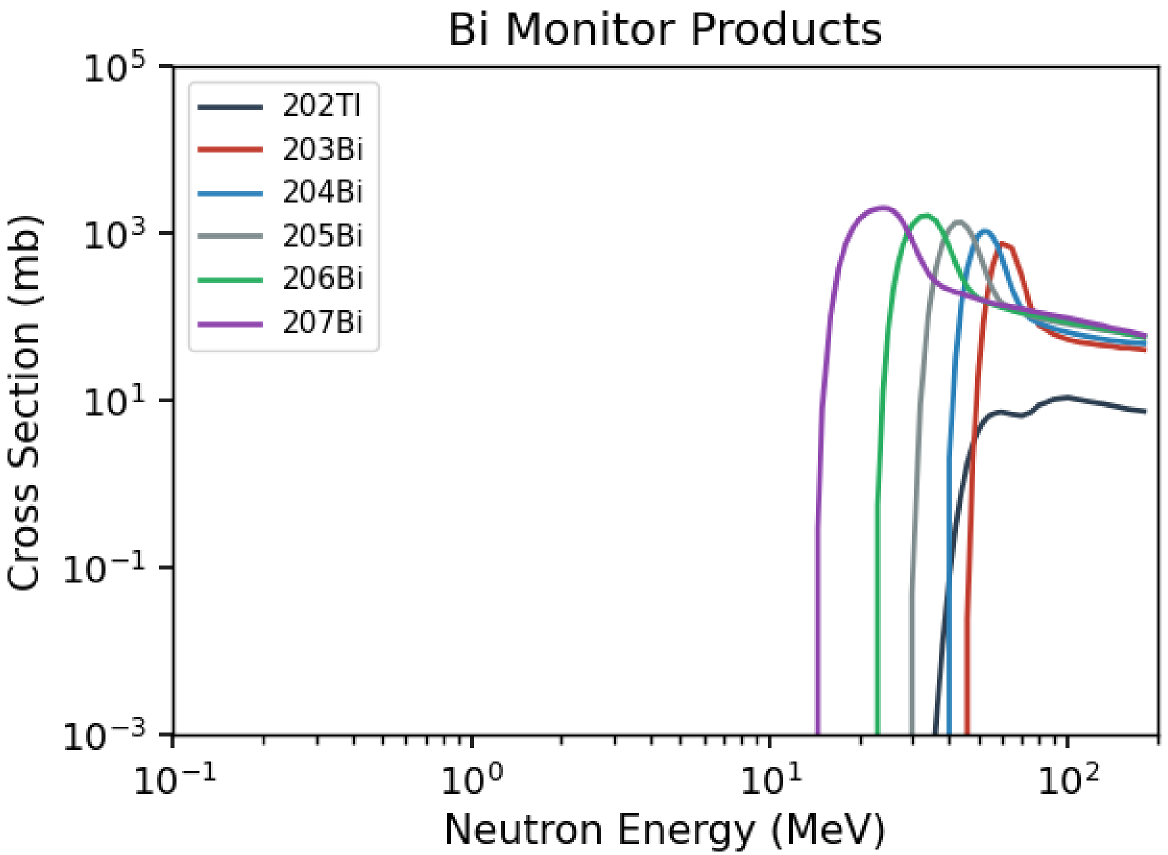
- Bi, Co, Ti foils chosen for irradiation
  - 25 mm x 25 mm, 0.25 mm thick
- 23 hour proton irradiation at 116 µA, 117 MeV, rastered beam
  - Time maximizes production of isotopes to 200 mCi control limit which includes short-lived isotopes (minute half-lives)
- Foils counted at BLIP after cooling period

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

Cross Sections for Monitor Isotopes

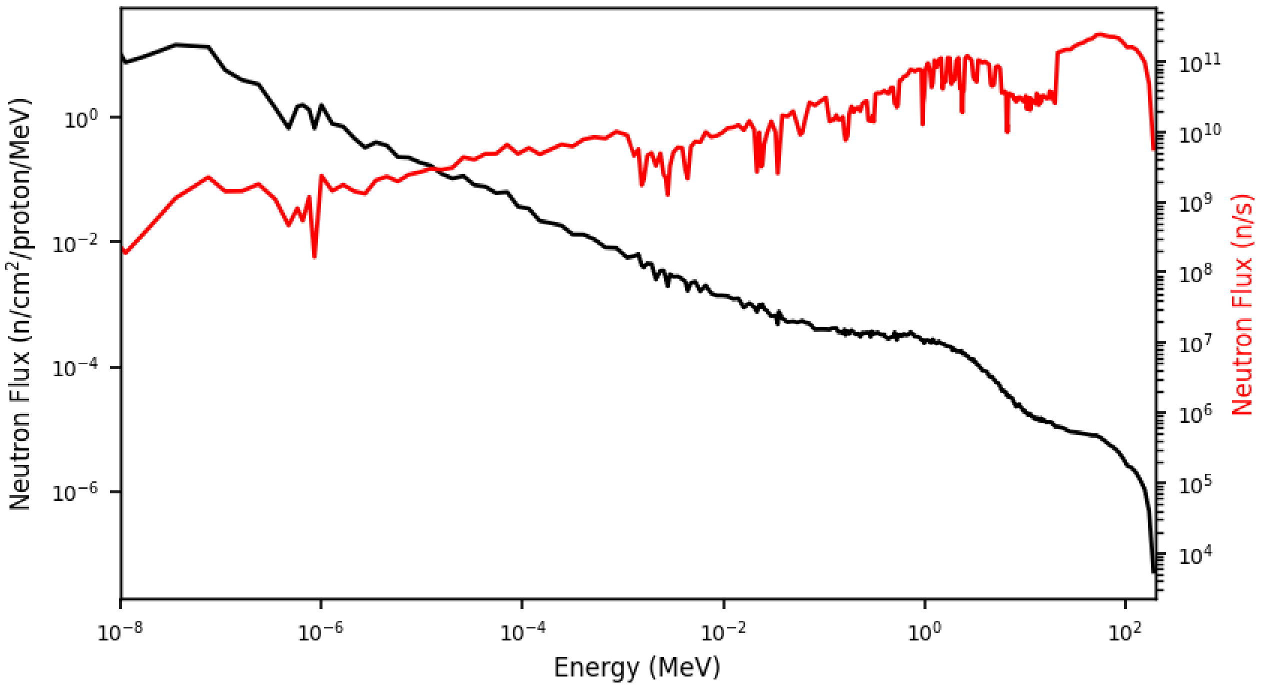




Neutron Spectrum Calculation

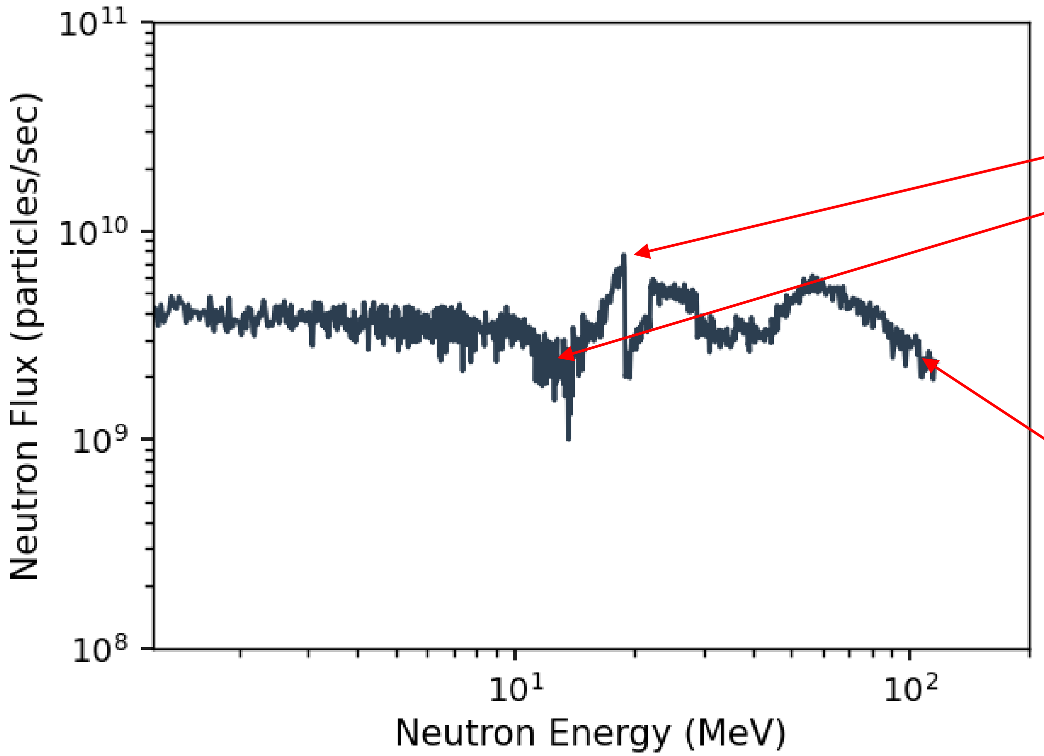
$$R = \int N \times \sigma(E) \times \Phi(E)$$

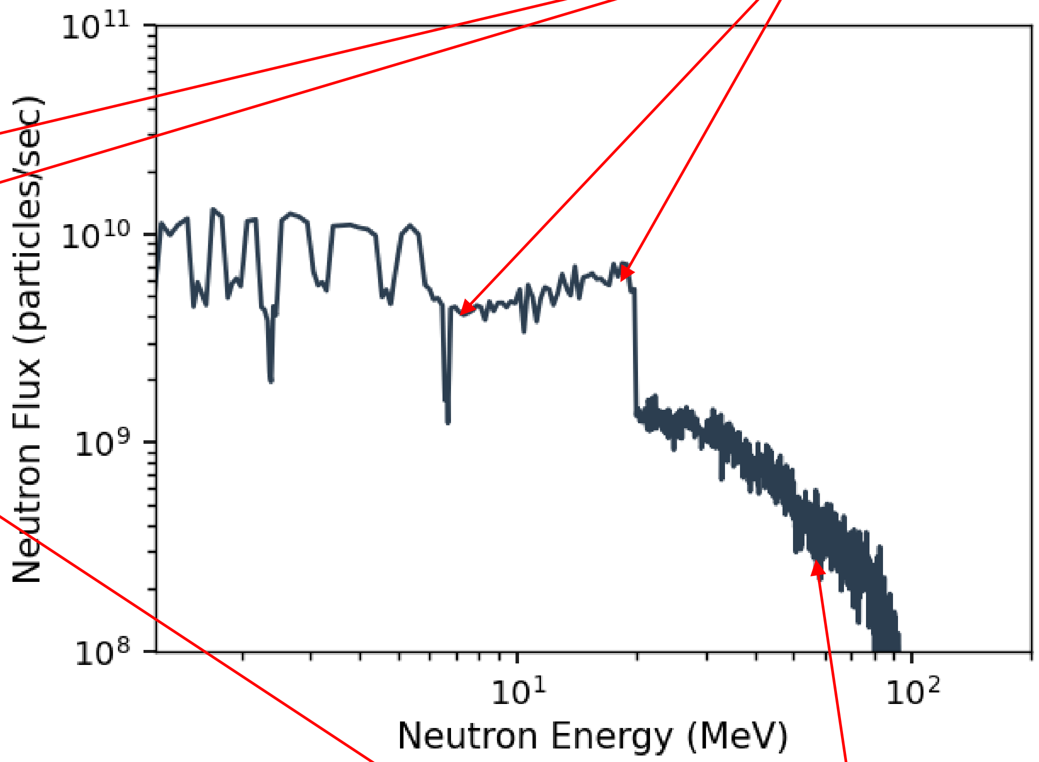
- Discrete energy points for cross section
- Cross sections from TENDL
- Monte Carlo variation of Φ(E)



Results of MC Calculation

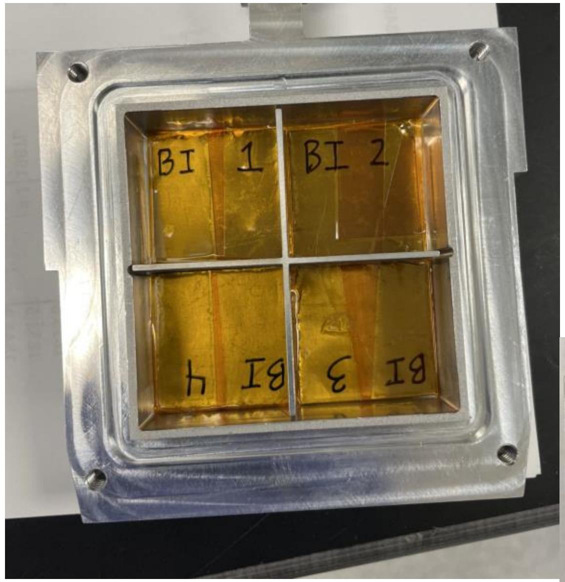
Similar features that correspond to nuclear data

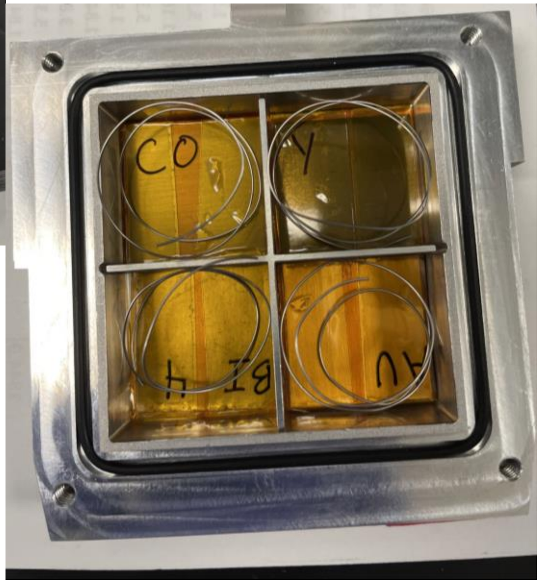




Other features may reflect low statistics or reliance on one or two cross sections

Irradiation – June 2022

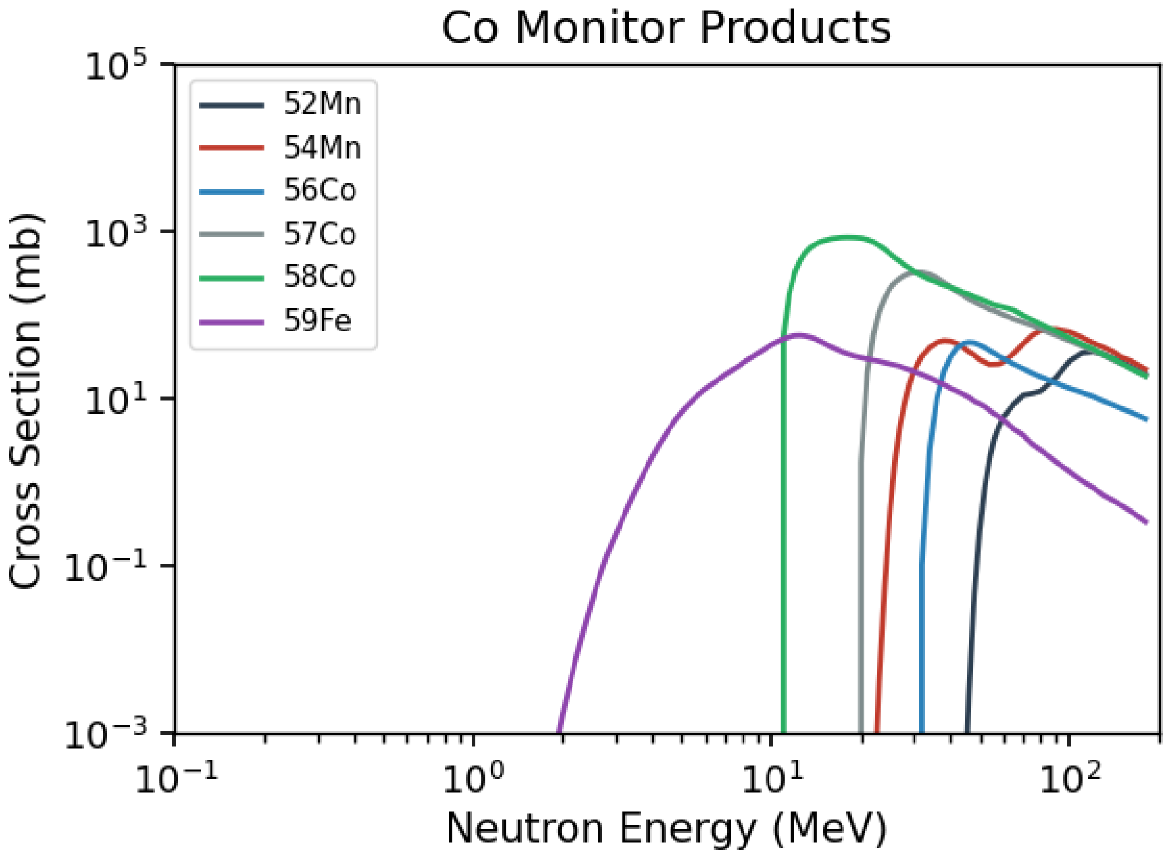


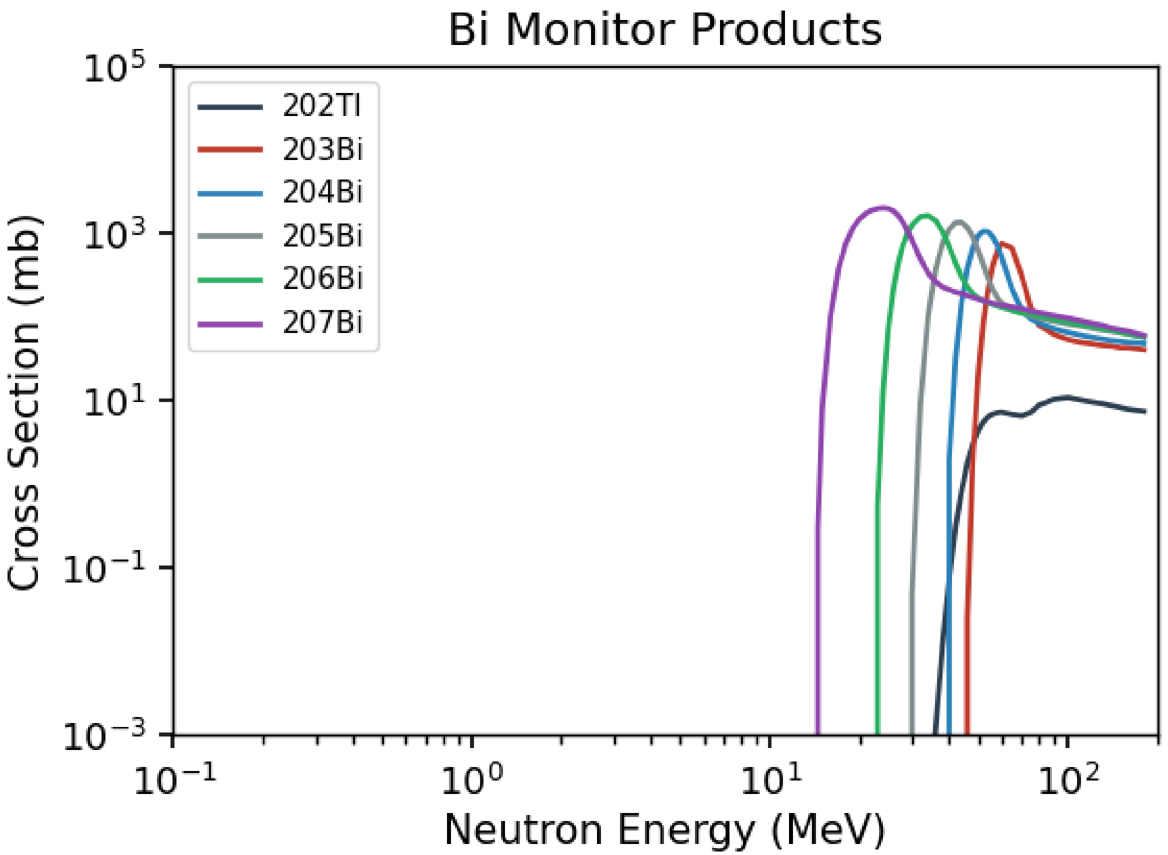


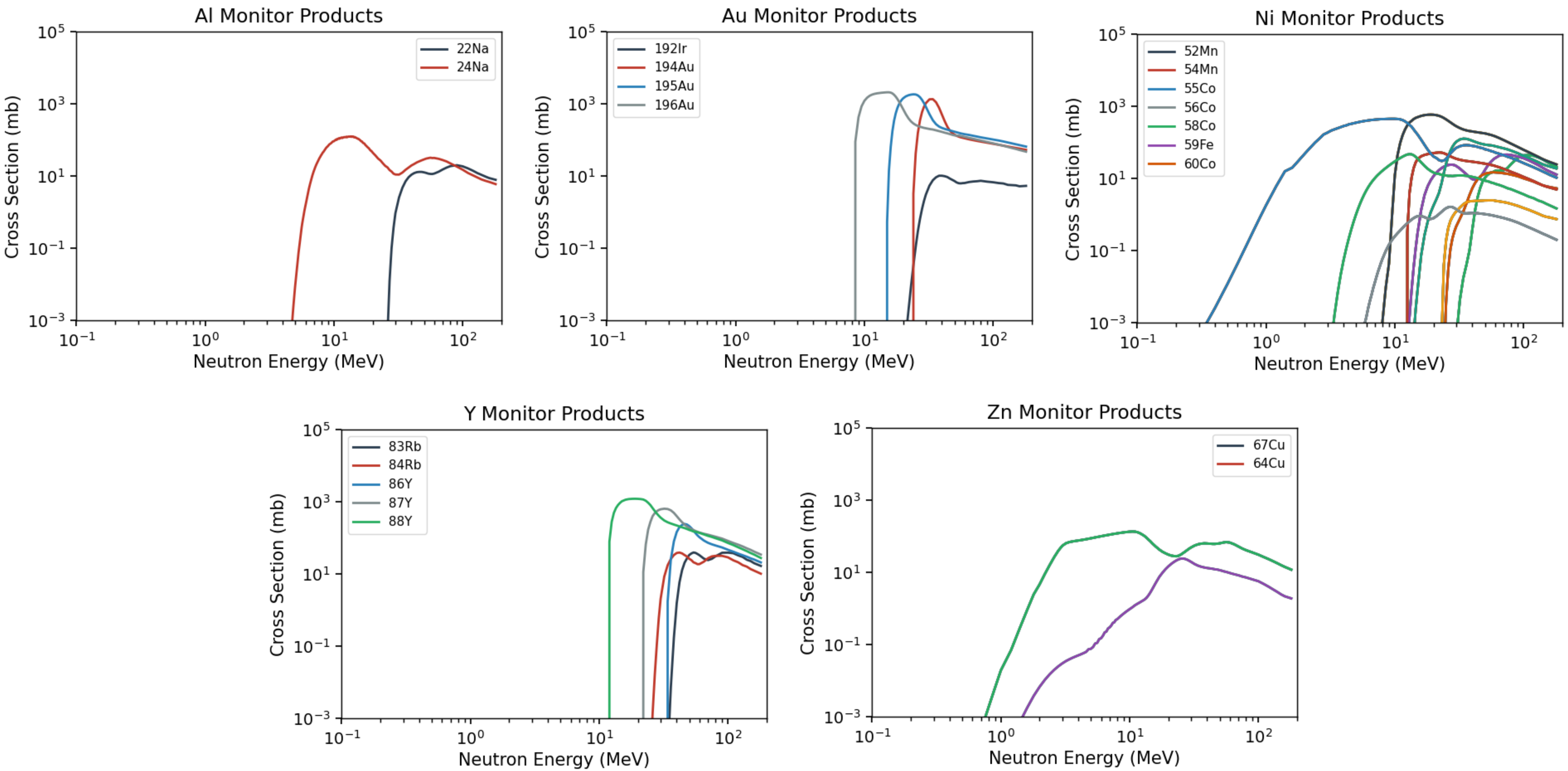
- 7 elements (natural abundance)

- 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


Co Monitor Products
52Mn
54Mn
56Co
57Co
58Co
59Fe
Cross Section (mb)
Neutron Energy (MeV)

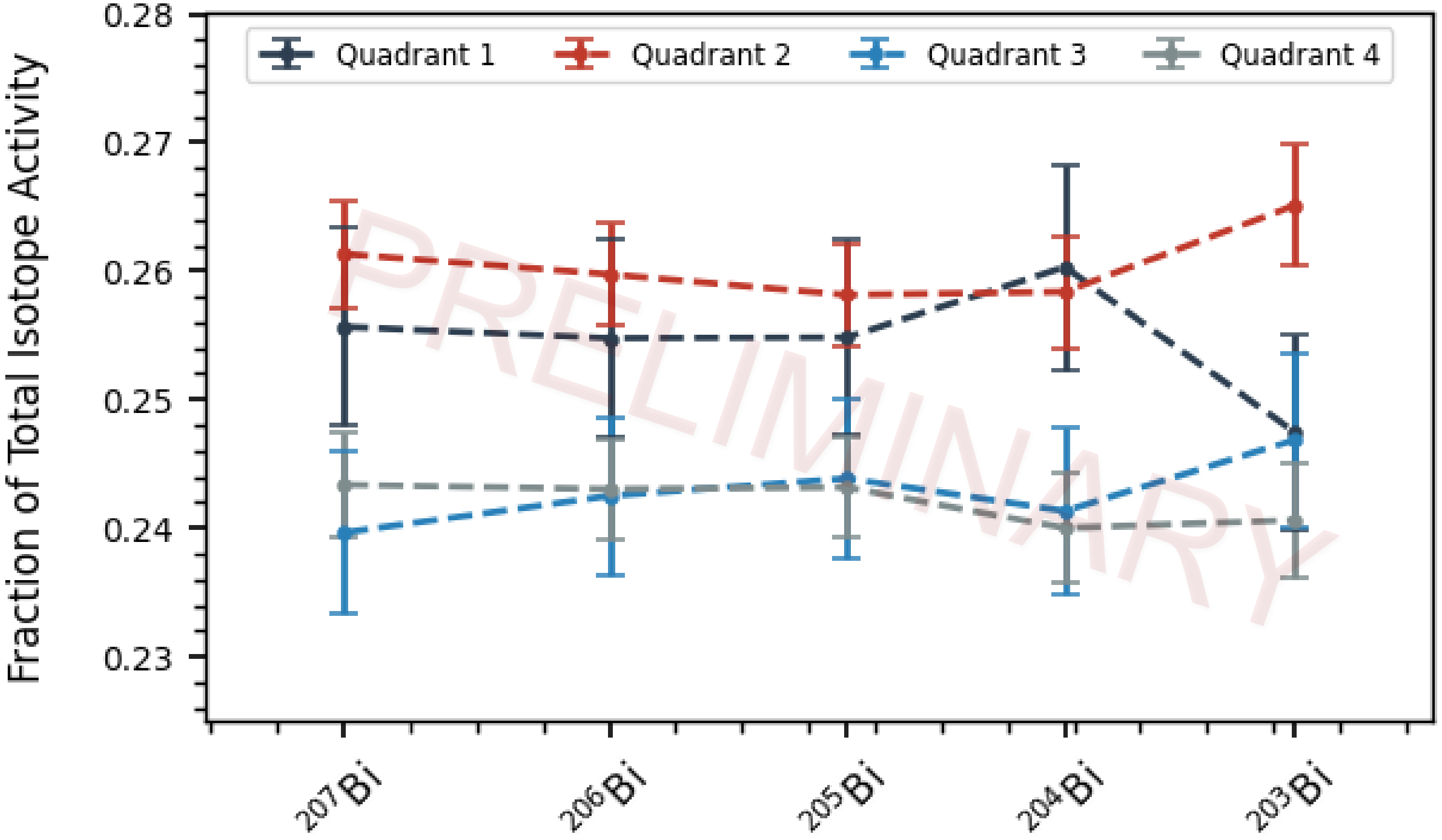

Bi Monitor Products
202Tl
203Bi
204Bi
205Bi
206Bi
207Bi
Cross Section (mb)
Neutron Energy (MeV)


Al Monitor Products
22Na
24Na
Au Monitor Products
192Ir
194Au
195Au
196Au
Ni Monitor Products
52Mn
54Mn
55Co
56Co
58Co
59Fe
60Co
Y Monitor Products
83Rb
84Rb
86Y
87Y
88Y
Zn Monitor Products
67Cu
64Cu
Cross Section (mb)
Neutron Energy (MeV)

New Products

Radionuclide	$t_{1/2}$ (days)	Activity
Zn Foil		
^{64}Cu	0.5	449.13 ± 3.45 µCi
^{67}Cu	2.6	12.62 ± 0.04 µCi
Y Foil		
^{86}Y	0.6	77.20 ± 0.14 µCi
^{88}Y	106.6	2.03 ± 0.01 µCi
Au Foil		
^{194}Au	1.6	163.61 ± 1.24 µCi
^{196}Au	6.2	68.69 ± 0.34 µCi

Quadrant Variation



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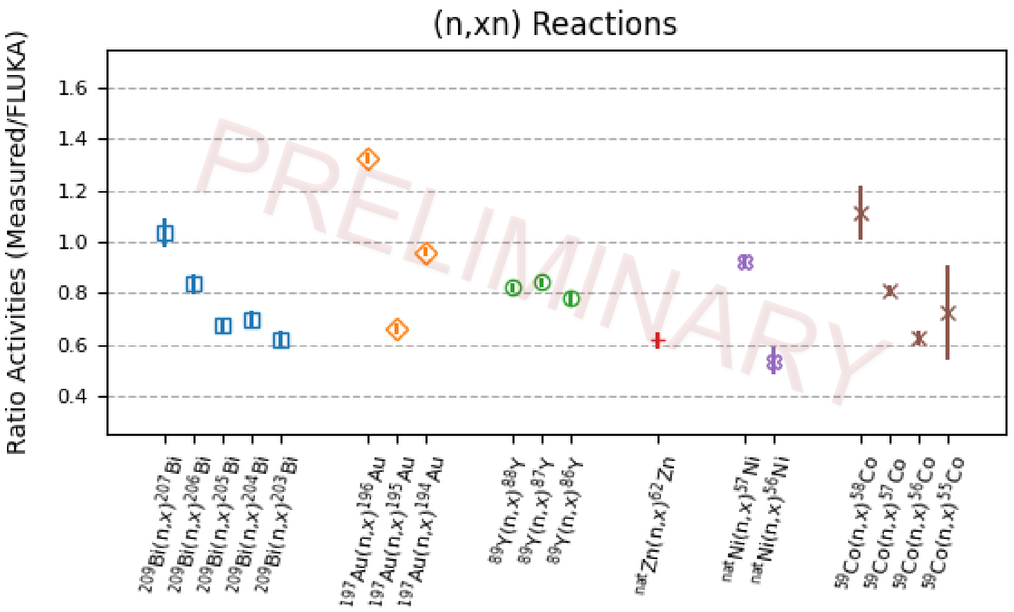
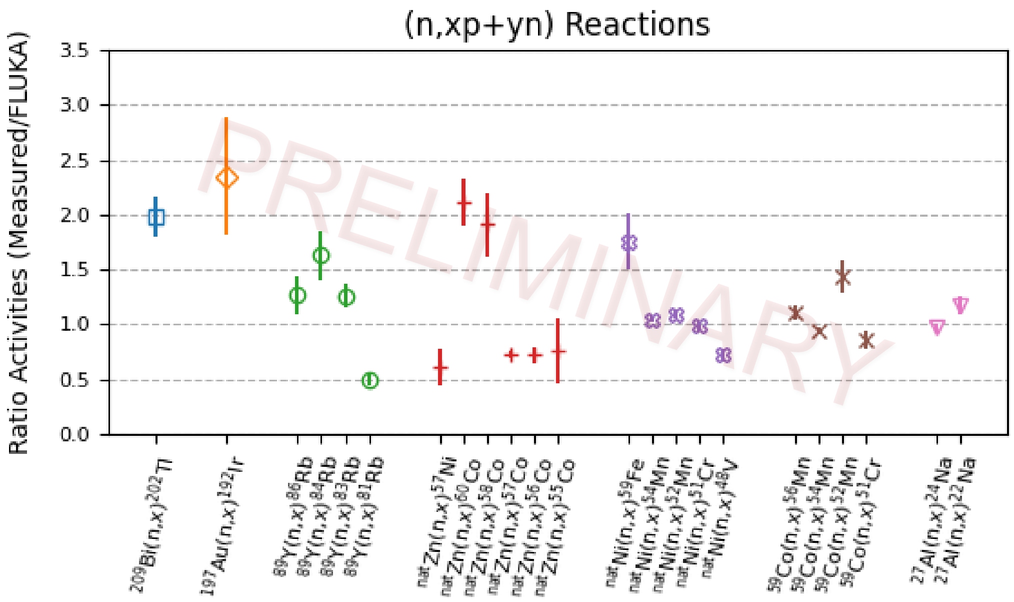
Results


(n,xn) Reactions
Ratio Activities (Measured/FLUKA)
1.6
1.4
1.2
1.0
0.8
0.6
0.4
PRELIMINARY
^{209}Bi(n,x)^{207}Bi
^{209}Bi(n,x)^{206}Bi
^{209}Bi(n,x)^{205}Bi
^{209}Bi(n,x)^{204}Bi
^{209}Bi(n,x)^{203}Bi
^{197}Au(n,x)^{196}Au
^{197}Au(n,x)^{195}Au
^{197}Au(n,x)^{194}Au
^{89}Y(n,x)^{88}Y
^{89}Y(n,x)^{87}Y
^{89}Y(n,x)^{86}Y
^{nat}Zn(n,x)^{62}Zn
^{nat}Ni(n,x)^{57}Ni
^{nat}Ni(n,x)^{56}Ni
^{59}Co(n,x)^{58}Co
^{59}Co(n,x)^{57}Co
^{59}Co(n,x)^{56}Co
^{59}Co(n,x)^{55}Co

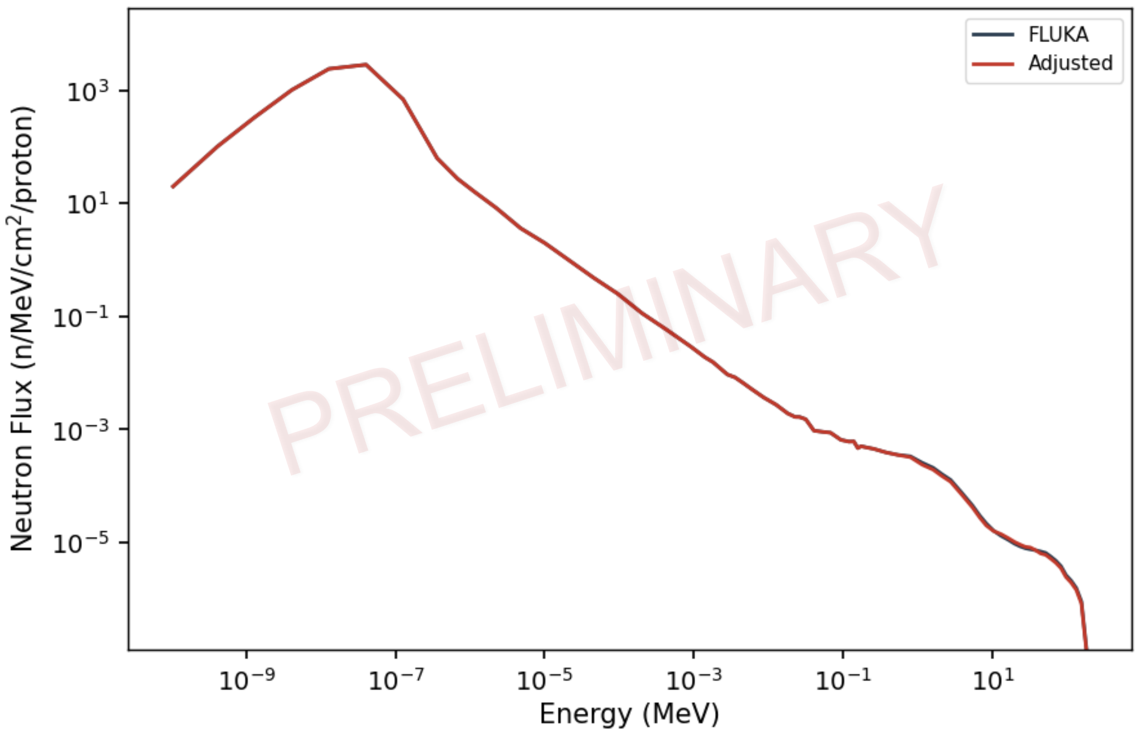

(n,xp+yn) Reactions
Ratio Activities (Measured/FLUKA)
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
PRELIMINARY
^{209}Bi(n,x)^{202}Tl
^{197}Au(n,x)^{192}Ir
^{89}Y(n,x)^{86}Rb
^{89}Y(n,x)^{84}Rb
^{89}Y(n,x)^{83}Rb
^{89}Y(n,x)^{81}Rb
^{nat}Zn(n,x)^{57}Ni
^{nat}Zn(n,x)^{60}Co
^{nat}Zn(n,x)^{58}Co
^{nat}Zn(n,x)^{57}Co
^{nat}Zn(n,x)^{56}Co
^{nat}Zn(n,x)^{55}Co
^{nat}Ni(n,x)^{59}Fe
^{nat}Ni(n,x)^{54}Mn
^{nat}Ni(n,x)^{52}Mn
^{nat}Ni(n,x)^{51}Cr
^{nat}Ni(n,x)^{48}V
^{59}Co(n,x)^{56}Mn
^{59}Co(n,x)^{54}Mn
^{59}Co(n,x)^{52}Mn
^{59}Co(n,x)^{51}Cr
^{27}Al(n,x)^{24}Na
^{27}Al(n,x)^{22}Na

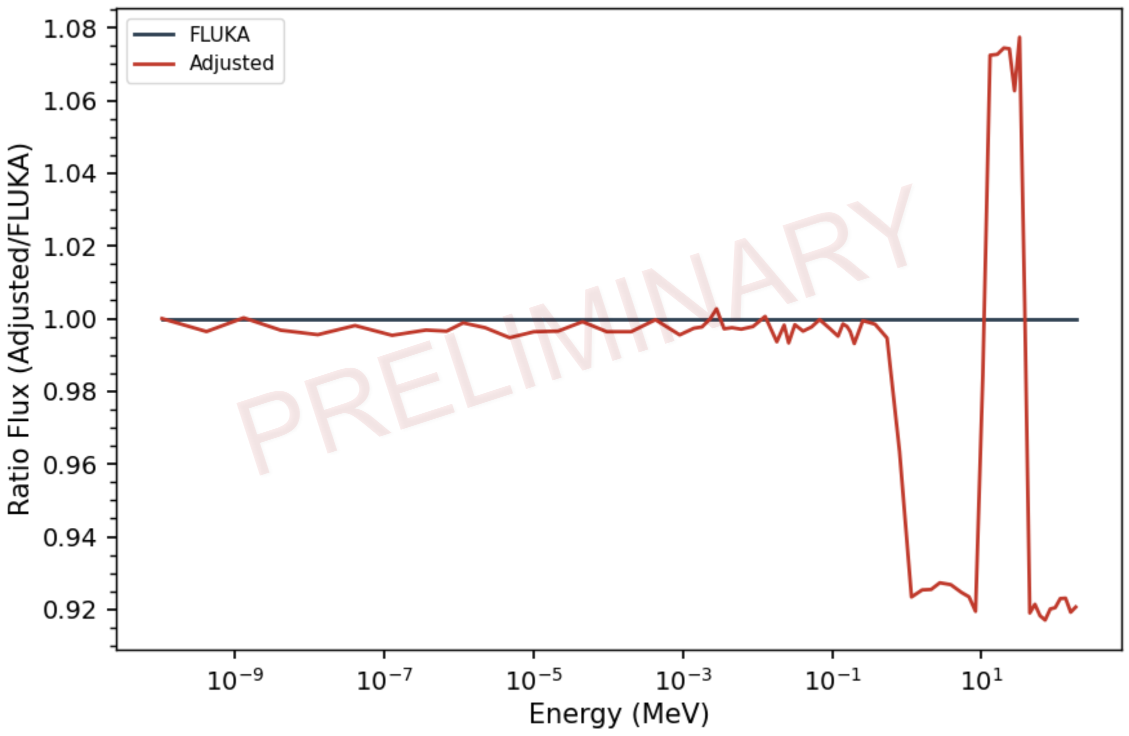
Radionuclide Production

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		0.77		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

Spectrum Adjustment





Summary

- Neutrons provide a way of producing additional radioisotopes that may be accessible through other means, and are already being generated at BLIP

- 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
    - Henryk Chelminski
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  - Radiation Control Division
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    - Vicky Litton
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