Low and Medium Energy Cyclotron Project Update

Pavithra Kankanamalage, Dohyun Kim, David Schlyer, Cathy Cutler

PI for the project: Jasmine Hatcher LaMarre

January 23, 2023

@BrookhavenLab

Outline

- Introduction
- Cross section
- FLUKA results
- Target development and separation

Introduction

- Purpose: examine potential production pathways for radionuclides that can be produced using low and medium-energy cyclotrons

- The list of radionuclides that can be produced using a beam ≤30MeV is extensive
  - isotopes of particular importance are Co-57, Cd-109, and Pb-203

- Plan to develop novel targets using methods such as electroplating and sputtering
  - these techniques will produce high-purity targets and avoid metal impurities

- Intend to test these targets using the BLIP and the 19 MeV MIRP cyclotron, evaluate separation methods, and determine the quality control requirements of these isotopes

Co-57, Pb-203, and Cd-109

Co-57

- Co-57 is commonly used in both industrial and medical practices as a calibration source
- The domestic and foreign supplies of Co-57 have halted due to company restructuring and geopolitical issues
- Using a cyclotron, Co-57 can be produced in high yield from a natural nickel target

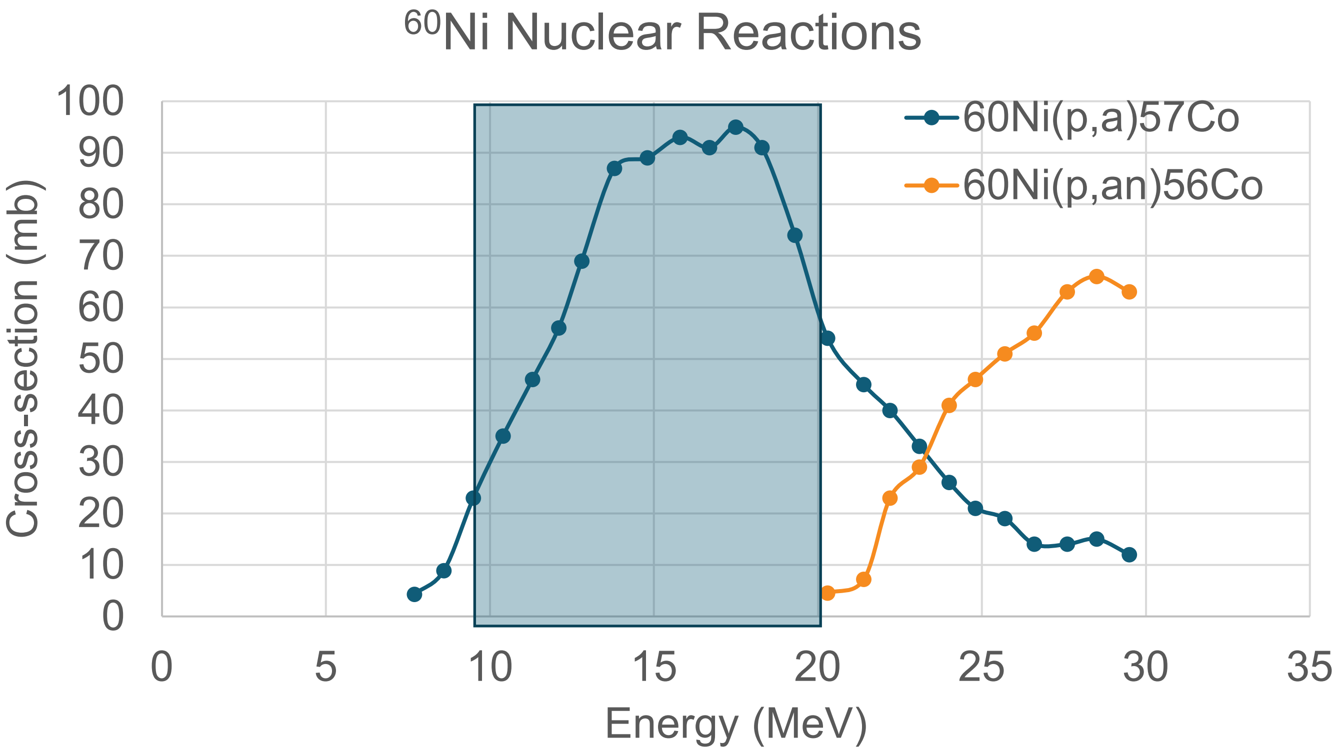
Pb-203

- Pb-203 is an isotope of interest as an imaging agent as well as part of a theragnostic pair with Pb-212
- The supply of Pb-203 has also halted
- Pb-203 can be produced in high yield from a natural or enriched thallium target

Cd-109

- Cd-109 can be used as a calibration source of X-ray and γ-ray systems as well as a tracer for Cd, which is an environmental pollutant
- Produced at the Isotope Production Facility at Los Alamos National Laboratory through the proton bombardment of an indium target

Cross-section



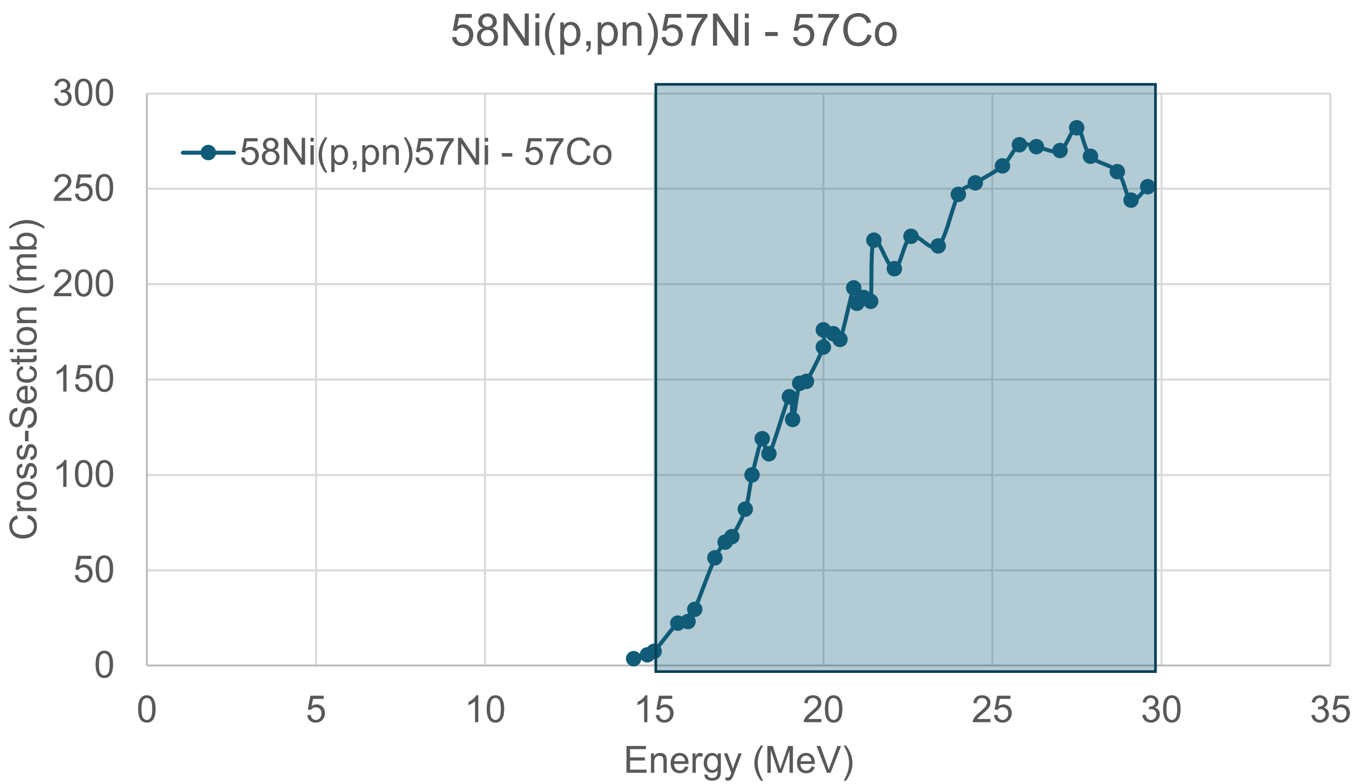
Range at 20 MeV is 756 microns
Range at 10 MeV is 231 microns

The target should be 525 microns thick

Ni-60 is 26.223 % Natural abundance

Business sensitive

Cross-section

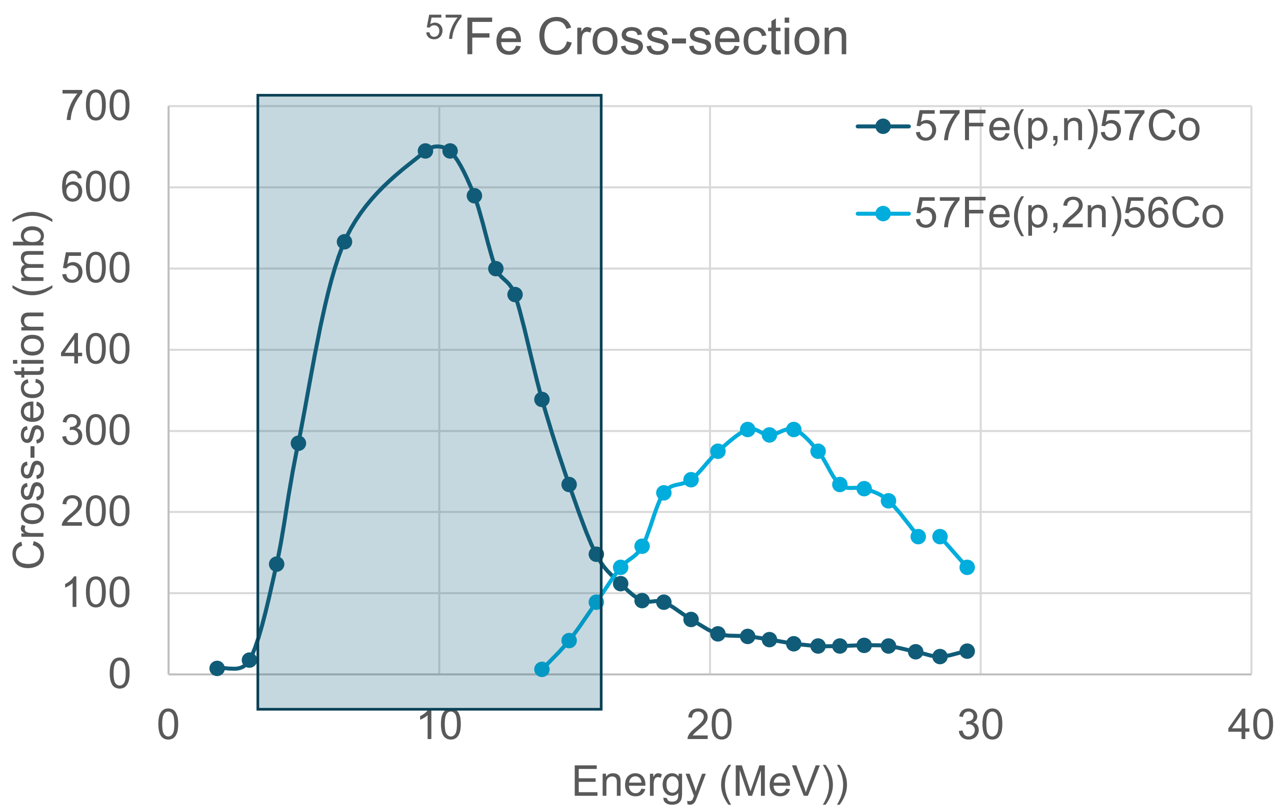


Range at 30 Mev is 1530 microns
Range at 15 MeV is 460 microns

The target should be 460 microns thick 1070 microns or 1 mm thick

Ni-58 is 68.077 %
Natural abundance

Cross-section



Range at 15 Mev is 516 microns
Range at 4 MeV is 56 microns

The target should be 460 microns thick

Fe-57 is 2.200 % Natural abundance

Brookhaven National Laboratory

Business sensitive

FLUKA DATA

- Ni58 target
  - 19 MeV, 200 uA, 40 hours irradiation, 0.065 cm thick and 1 cm diameter

A	Sym.	Z	Bq	Err.	T 1/2 (s)	Ci
55	Co	27	5.31E+10	+/- 0.4 %	6.31E+04	1.43E+00
57	Co	27	4.10E+09	+/- 0.2 %	2.35E+07	1.11E-01
57	Ni	28	7.38E+10	+/- 0.3 %	1.28E+05	1.99E+00
58	Cu	29	1.97E+11	+/- 0.4 %	3.20E+00	5.33E+00

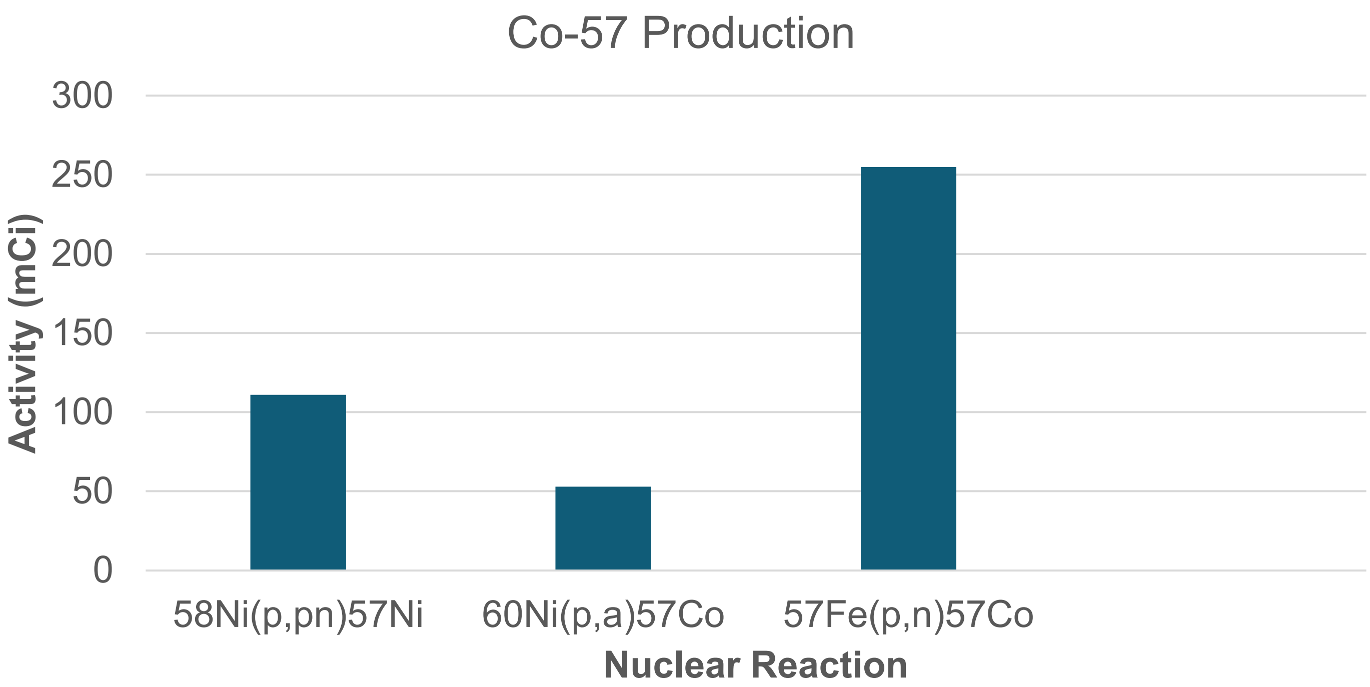
- Ni60 target
  - 19 MeV, 200 uA, 40 hours irradiation, 0.07 cm thick and 1 cm diameter

A	Sym.	Z	Bq	Err.	T 1/2 (s)	Ci
57	Co	27	1.97E+09	+/- 0.2 %	2.35E+07	5.33E-02
59	Cu	29	1.54E+09	+/- 2.4 %	8.15E+01	4.16E-02
60	Cu	29	1.17E+12	+/- 0.1 %	1.42E+03	3.16E+01

- Fe57 target
  - 19 MeV, 200 uA, 40 hours irradiation, 0.052 cm thick and 1 cm diameter

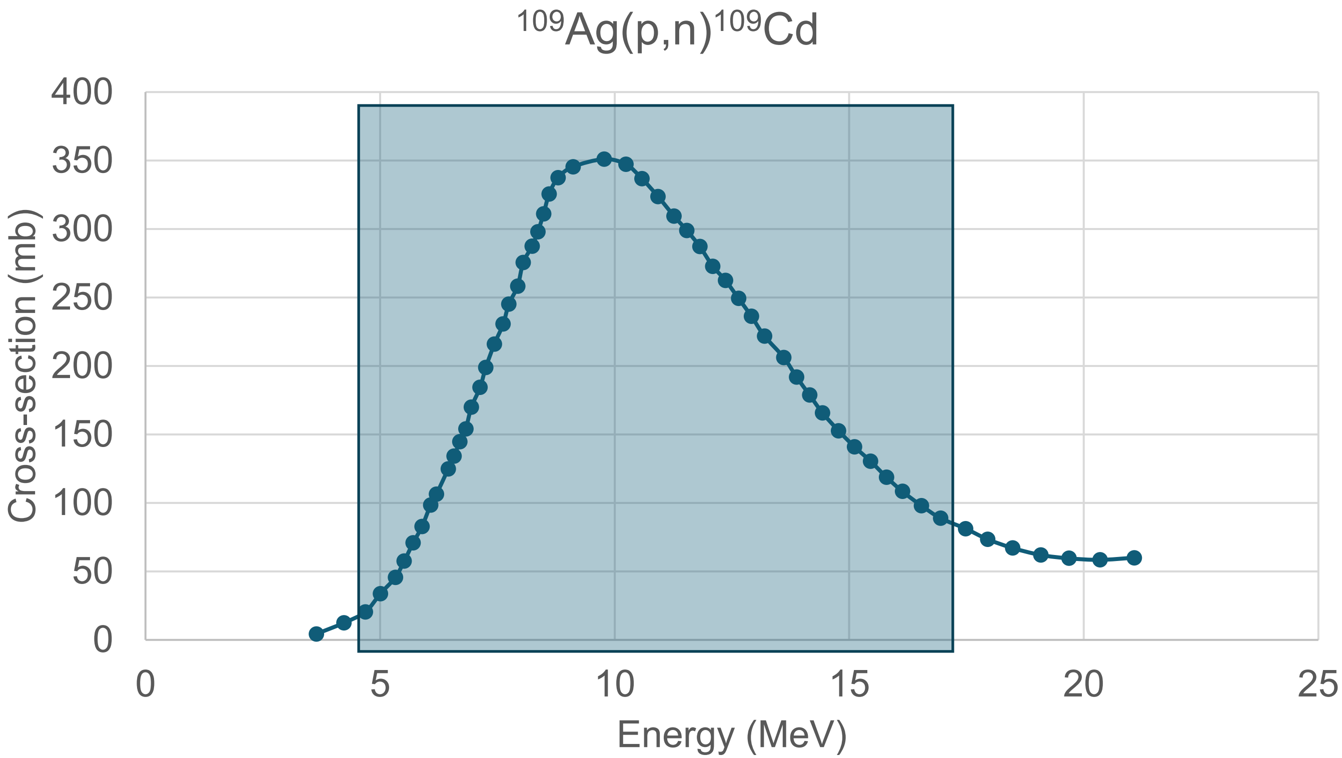
A	Sym.	Z	Bq	Err.	T 1/2 (s)	Ci
54	Mn	25	7.20E+08	+/- 0.3 %	2.70E+07	1.94E-02
56	Co	27	4.17E+08	+/- 0.8 %	6.68E+06	1.13E-02
57	Co	27	9.44E+09	+/- 0.1 %	2.35E+07	2.55E-01

Comparison of Production Routes for Co-57



Ni-58 – 68% natural abundance
Ni-60 – 26% natural abundance
Fe-57 – 2.2% natural abundance

Cross-section



Range at 17 MeV
600 microns

Range at 5 MeV
80 microns

Thickness
520 microns

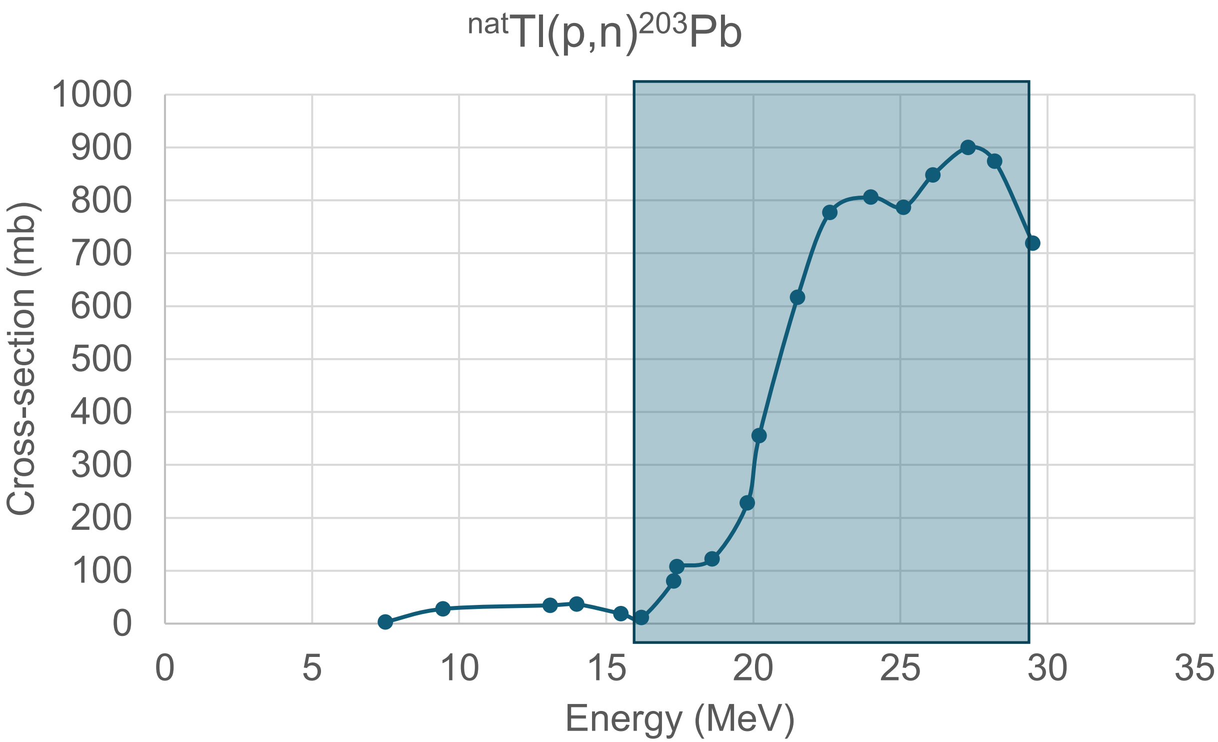
Natural Abundance
48%

FLUKA DATA

- Ag target
  - 19 MeV, 200 uA, 40 hours irradiation, 0.0724 cm thick and 1 cm diameter

A	Sym.	Z	Bq	Err.	T 1/2 (s)		Ci
106	Ag	47	1.49E+10	+/- 1.0 %	1.44E+03		4.02E-01
107	Cd	48	5.49E+11	+/- 0.2 %	2.34E+04		1.48E+01
108	Ag	47	7.15E+09	+/- 1.0 %	1.42E+02		1.93E-01
110	Ag	47	1.50E+09	+/- 1.1 %	2.46E+01		4.04E-02
Isomers							
A	Sym.	Z	mth	Bq	Err.	T 1/2 (s)	
106	Ag	47	1	1.90E+09	+/- 1.0 %	7.31E+05	5.13E-02
107	Ag	47	1	6.05E+11	+/- 0.2 %	4.43E+01	1.64E+01
109	Ag	47	1	4.74E+10	+/- 0.5 %	3.96E+01	1.28E+00
109	Cd	48	1	9.46E+10	+/- 0.3 %	1.20E-05	2.56E+00
109	Cd	48	2	9.46E+10	+/- 0.3 %	1.09E-05	2.56E+00

Cross-section



Range at 28 MeV is 1600 microns

Range at 16 MeV is 618 microns

Target thickness is 982 microns

Natural Abundance 29.5%

FLUKA DATA

- Tl target
  - 30 MeV, 200 uA, 40 hours irradiation, 0.176 cm thick and 1 cm diameter

A	Sym.	Z	Bq	Err.	T 1/2 (s)		Ci
200	Tl	81	6.44E+10	+/- 1.1 %	9.40E+04		1.74E+00
200	Pb	82	1.43E+11	+/- 1.1 %	7.74E+04		3.87E+00
201	Tl	81	3.10E+11	+/- 0.2 %	2.63E+05		8.37E+00
201	Pb	82	1.30E+12	+/- 0.2 %	3.36E+04		3.50E+01
202	Tl	81	1.32E+10	+/- 0.7 %	1.06E+06		3.57E-01
203	Pb	82	1.39E+12	+/- 0.1 %	1.87E+05		3.75E+01
Isomers							
A	Sym.	Z	mth	Bq	Err.	T 1/2 (s)	
201	Pb	82	1	6.86E+11	+/- 0.3 %	6.10E+01	1.85E+01
202	Pb	82	1	9.97E+11	+/- 0.2 %	1.27E+04	2.69E+01
203	Pb	82	1	1.12E+12	+/- 0.2 %	6.30E+00	3.02E+01
203	Pb	82	2	1.12E+12	+/- 0.2 %	4.80E-01	3.02E+01
204	Pb	82	1	1.04E+12	+/- 0.2 %	4.03E+03	2.81E+01
205	Pb	82	1	7.78E+10	+/- 1.0 %	5.54E-03	2.10E+00

Target Development and Separation

- Co-57
  - Ni electroplated Au substrate
  - AG1-X8 anion-exchange resin from Bio-Rad laboratories can be used to separate Co from Ni
- Cd-109
  - Ag electroplated Cu substrate
  - Purification by precipitating the AgCl and further purifying the Cd with AG1-X8 anion-exchange resin from Bio-Rad laboratories
- Pb-203
  - Tl electroplated Cu substrate
  - Purification with Pb resin from Eichrom Technologies (Lisle, IL)

Ref: 1. Mirzaee, M., Sadeghi, M., Gholamzadeh, Z., & Lahouti, S. (2008). Thick silver electrodeposition on copper substrate for ^{109}Cd production, *J. Radioanal. Nucl. Chem.*, *277*(3), 645-650 (2008)

2. Yarmohammadi, M., Mirzaei, M. & Samadi-Maybodi, A. Application of Ion Exchange, Solvent Extraction, and Ion-Imprinted Methods for Separation of ^{203}Pb. *Radiochemistry* **61**, 724–727 (2019).

3. McNeil, B.L., Robertson, A.K.H., Fu, W. et al. Production, purification, and radiolabeling of the 203Pb/212Pb theranostic pair. EJNMMI radiopharm. chem. 6, 6 (2021).

4. Sadeghi, M., Karami, H., Sarabadani, P. *et al.* Separation of the no-carrier-added ^{109}Cd from Ag, Cu and ^{65}Zn by use of a precipitation and AG1-X8 resin. *J. Radioanal. Nucl. Chem.*, **281**, 619 (2009).

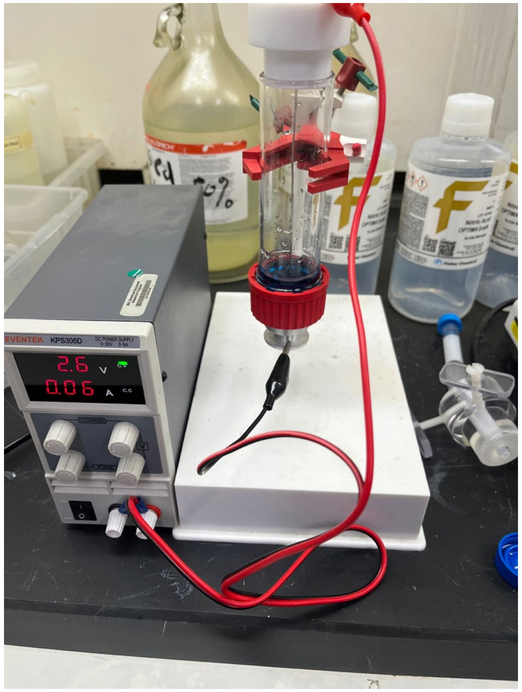
5. Barrett KE, Houson HA, Lin W, Lapi SE, Engle JW. Production, Purification, and Applications of a Potential Theranostic Pair: Cobalt-55 and Cobalt-58m. Diagnostics (Basel). 11(7):1235 (2021)

Preliminary Data for Ni Electrodeposition

- Cathode: Gold
- Anode: Platinum
- Potential applied: 2.6 V
- Duration: 16 h
- Component of the solution:
    - $NiSO_4.XH_2O$ (462 mg) and $(NH_4)_2SO_4$ (300 mg),
    - 20 mL solution volume
    - pH 9

$NiSO_4.XH_2O$ (462 mg) + $(NH_4)_2SO_4$ (300mg) in water

$[Ni(NH_3)_6]^{2+}$ complex after adjusting the pH to 9



Electrochemical cell

Ref: Piel H, Qain S.M, Stücklin G. Excitation functions of (p,xn)-reactions on ^{nat}Ni and highly enriched ^{62}Ni: possibility of production of medically important radioisotope ^{62}Cu at a small cyclotron. Radiochim Acta. 1992;57(1):1–6.

Brookhaven National Laboratory

Business sensitive

Preliminary Data for Ni Electrodeposition

- Assuming $NiSO_4.XH_2O$ is $NiSO_4.6H_2O$

The Ni mass of the electroplating solution = 103.2 mg

The Ni mass deposited on the Au plate =101.4 mg

Yield = 98.3%

- ICP-OES analysis of the solutions underway to determine the yield.

- Future studies:
  - SEM analysis to determine the homogeneity of the deposition
  - SEM-EDS analysis to determine the composition of the Ni deposit

Solution after 16 h electrolysis

Electrodeposited Ni on a Gold plate

Ni sediments settled on the Pt Anode

Summary

- We are working on producing Co-57, Cd-109, and Pb-203 using low and medium energy cyclotron
- We have compared the Cross-section data and performed FLUKA analysis of the production of Co-57, Cd-109, and Pb-203
- Ni electrodeposition on an Au substrate has been demonstrated successfully to use as a target for Co-57 production