

Effects of Neutron Depression on the Specific Activity of Ni-63

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Acknowledgments

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Abstract

The objective of this thesis is to investigate the effects of neutron flux depression on the specific activity Ni-63. Ultimately, the goal of this experiment is to verify a method for creating a higher yield of any desired nuclide produced in a reactor while simultaneously requiring less target material. This experiment was implemented by loading two stacks of highly enriched Ni-62 foils into the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL) and analyzing variations in specific activity in the foils based on their position. In order to ensure accurate results, the samples were chemically treated to remove all radioimpurities via a series of ion exchange columns. The experimental results were also compared to simulated results obtained by an analytical program called IsoChain and the Monte Carlo based simulation program, MCNP.

Table of Contents

Chapter 1: Background	1
1.1 ^{63}Ni Description	1
1.2 ^{63}Ni Production	1
1.3 Detection	2
1.4 Commercial Uses	3
1.5 Neutron Depression	3
1.5.1 Neutron Interactions	3
1.5.2 Flux Depression	4
Chapter 2: Project Objectives	5
2.1 Project Description	5
2.2 Target Description	8
2.2.1 Foil Composition	8
2.2.2 Foil Geometry and Positioning	8
Chapter 3: Theory and Methodology	11
3.1 Foil Dissolution	11
3.2 Sample Dilution and Liquid Scintillation Counting	14
3.3 Experimental Calculations and Error Analysis	18
3.4 Liquid Chromatography	20
3.4.1 Column Construction	20
3.4.2 Co-60 Removal (MP1 Column)	21
3.4.3 Eu-152 Removal (AG50x8 Column)	22
3.4.4 Post-Separation Processing	25
3.4.5 Error Corrections	28
3.5 IsoChain Theory	29
3.6 MCNP6.2 Theory	32

3.6.1 MCNP Structure.....	32
3.6.2 Experiment Specifications	33
Chapter 4: Results.....	36
4.1 Pre-Separation Results	36
4.2 Post Separation Results.....	36
4.3 Isochain Comparisons	41
4.4 MCNP Comparison.....	42
Chapter 5: Conclusions	44
References	46
Appendices	48
Appendix A: IsoChain Calculations	49
Appendix B: Foil LSC Measurements	50
Appendix C: MCNP Input	52
Vita	53

List of Tables

Table 1: Target NM-901 Rabbit Data	10
Table 2: Foil Weights and Isotopic Distribution.....	12
Table 3: IsoChain Case Specifications	30
Table 4: IsoChain Cross-Section Data	31
Table 5: Post-Separation vs. Pre-Separation Comparison.....	39
Table 6: Pre-Separation Experimental vs. Isochain Results.....	41
Table 7: Post-Separation Experimental vs. Isochain Results	42
Table A.1: IsoChain Calculated Sample Activity	49
Table B.1: Pre-Separation LSC Measurements.....	50
Table B.2: Post-Separation LSC Measurements and Calculations: A1-A6	50
Table B.3: Post-Separation LSC Measurements and Calculations: B1-B6.....	51

List of Figures

Figure 1: Nickel foil's orientation inside Hydraulic Tube "Rabbit"	6
Figure 2: Target flux profile estimation	7
Figure 3: Enriched Nickel Foil Assay	9
Figure 4: Nickel Foils Dimensions	10
Figure 5: Nickel Foils in HPGe Counting Cards	12
Figure 6: Characteristic Green of Dissolved Nickel Foil	13
Figure 7: Sample Evaporation Setup.....	15
Figure 8: Hidex Liquid Scintillation Counter	16
Figure 9: Example Quenching Curve	17
Figure 10: MP1 Column	23
Figure 11: AG50x8 Column	24
Figure 12: HPGe Setup	26
Figure 13: Gamma Spectra during Chemical Separation	26
Figure 14: Chemical Separation Diagram.....	28
Figure 15: IsoChain Input Screen	31
Figure 16: MCNP Geometries Snapshots	35
Figure 17: Specific Activity of Samples A1-A6 before Chemical Separation	37
Figure 18: Specific Activity of Samples B1-B6 before Chemical Separation.....	37
Figure 19: Specific Activity of Samples A1-A6 after Chemical Separation	38
Figure 20: Specific Activity of Samples B1-B6 after Chemical Separation.....	38
Figure 21: Average Specific Activities between Foil Sets	40
Figure 22: MCNP Cell Flux Values for Lower Target	43
Figure 23: MCNP Cell Flux Values for Lower Target	43

List of Attachments

File 1: Mughabghab, 2006 (Mughabghab 2006.xlsx)

File 2: NM-901 Foil Geometry Sheet (20180621070157369.pdf)

File 3: NM-901 EXCITE Irradiation History (NM-901 irradiation history.pdf)

File 4: Eckert and Zeigler ^{63}Ni Standard Datasheet (Certificate of Calibration for Beta Standard Ni63 Source #1835-13-1.pdf)

File 5: MCNP Model (HFIR2_Ni.txt)

File 6: Raw Pre-Separation Data (ZW Ni-63 QC PRE-SEP 4-9-19.xlsx)

File 7: Raw Post-Separation Data (ZW Ni-63 PS Quenching Curve 4-9-19.xlsx)

Chapter 1: Background

1.1 ^{63}Ni Description

Nickel is a transition metal with an atomic number (Z) of 28 and naturally occurs as isotopes ^{58}Ni , ^{60}Ni , ^{61}Ni , ^{62}Ni , and ^{64}Ni . It is most commonly found in a Ni^{2+} oxidation state, but can also occur in compounds as Ni^0 , Ni^+ , and other experimentally produced states. ^{63}Ni itself is produced via neutron capture by ^{62}Ni . It has a half-life of 101.2 years and decays solely via beta decay to ^{63}Cu . The thermal neutron cross section of ^{62}Ni is 14.9 barns with a resonance integral of 0.3 barns, according to Mughabghab 2006 [ATTACHMENT 1]. As stated previously, once ^{63}Ni is created, it decays entirely through beta-minus (electron) emission with an energy of 66keV. This quality of being a pure, low energy beta-emitter makes ^{63}Ni both useful and potentially dangerous.

1.2 ^{63}Ni Production

In order to produce significant quantities of ^{63}Ni , natural nickel must first be enriched to contain a high concentration of ^{62}Ni . This can be achieved through a variety of methods, but the method applied for the enriched material used in this experiment was electromagnetic separation, employed by calutrons. Calutrons use the slight mass differences in the isotopes to purify a sample by ionizing the element and accelerating it via electric fields down a curved chamber and deflected by a magnetic field. The isotopes are accelerated evenly due to their identical charge, but the varying masses of the different isotopes cause the magnetic field to deflect them at varying angles. They are then deposited into varying bands along the plating of the chamber. [1] The High-Flux Isotope Reactor at Oak Ridge National Laboratory produces the highest reactor-based flux of neutrons as well as the one of the highest steady-state neutron fluxes of any research reactors in the United States. HFIR is used to produce industrial, research, and medical radioisotopes as well as study the effects of such a hostile environment on materials. One of the most unique features of HFIR is the hydraulic tubes, which allow researchers to insert and

remove material from the flux trap at any time. This is where the rabbit containing the ^{62}Ni foils used in this experiment was placed. [2]

1.3 Detection

Because ^{63}Ni does not emit gamma rays, some conventional methods for the detection of the isotope are ineffective. In fact, the only valid methods for detecting ^{63}Ni contamination are through gas filled counters or liquid scintillation.

Gas-filled counters, such as Geiger-Muller counters, utilize a device called an ionization chamber to detect the beta particles, as well as other charged particles. This is done by applying a potential difference between two plates, creating an electric field between them. When a charged particle, such as a beta particle, enters into this electric field, it slows down and ionize gas atoms within, ejecting electrons and leaving positive ions behind. Some of these ion pairs then drift into the circuit, creating an electrical current. The amount of ion pairs that reach the circuit is dependent on the voltage applied; therefore, once the voltage is increased to a certain point, which proportionally increases the electric field, all ion pairs are eventually captured. [3]

The method of detection for this thesis, however, is liquid scintillation. The mechanism of liquid scintillation is somewhat similar to that of crystal scintillation; however, it operates by dissolving or suspending active material in a special solution called the “cocktail” that is comprised of two main ingredients, a solvent and phosphors, or fluors, and possibly a surfactant to lower surface tension. The solvent is usually an aromatic organic compound due to its ability to not only dissolve the fluor, but efficiently collect energy from emission events, transfer this energy to the phosphor molecules rather than dissipating it by some other means, and also not quench the scintillation emitted by the fluors. The fluors, which only constitute 0.3-1% of the solution by volume, are comprised of primary scintillators, which accept energy from the solvent and produce an excited, light emitting state, and secondary scintillators, which shift the wavelength of the light emitted by the primary scintillator to a wavelength that is more easily counted by the photomultiplier tube. It also lessens the amount of reabsorption or the emitted photons by the primary scintillator material [3]

1.4 Commercial Uses

Explosives Detection - Most people have seen TSA agents at the airport swab luggage or other items of random passengers to check for explosives or narcotics. What most don't know is that the detector being used for these checks are Electron Capture Detectors that use ^{63}Ni as the radioactive source that ejects electrons into the air. If these electrons are captured by certain compounds, such as residue from explosives, the detector notes the decrease in electrons and alerts the security officers.

Surge Protectors - ^{63}Ni is also used in many modern ceramic cylinder surge protectors and voltage regulators. A thin layer of ^{63}Ni is placed in one electrode of the surge protectors and voltage regulators in order to ionize the fill gas of the cylinder, which prevent sudden transients of high voltage. [4]

BetaVoltaic Batteries - One of the most fascinating uses for ^{63}Ni is in betavoltaic batteries. Radioisotopes have been studied for some time now for their possible applications for small scale power sources due to their exceptionally long shelf life. The most popular example of these is radioisotope thermoelectric generators (RTG's), which have been used extensively for interstellar probes among other things. RTG's use an array of thermocouples to convert the heat created by the radioactive decay of radioisotopes into electricity. However, betavoltaic batteries use the beta particles (electrons) emitted by the source to create an electrical current. These have potential applications in the medical, military, and space exploration fields, and recent improvements to the batteries' designs make this technology very promising.

1.5 Neutron Depression

1.5.1 Neutron Interactions

During fission or radioactive decay, neutrons can be ejected from the nucleus and can then interact with other matter, specifically the nuclei of other nuclides. These interactions can be broken down into three categories: transfer, scattering, and absorption.

Transfer reactions typically occur at low energies and consist of one or more neutrons transferring from one particle to another; however, it is more likely that a scattering or absorption event will occur. Scattering events can be further broken down into two subcategories: elastic and inelastic. In an elastic scattering event, the total kinetic energy of the system is conserved, but in an inelastic scattering event, the target nucleus absorbs some of the kinetic energy, moving it into an excited state. The most important interaction for this experiment, however, is neutron capture/absorption. This occurs when a free neutron collides with the nuclei of an atom and joins to it, making the nucleus heavier and pushing it to an excited state. This typically results in the immediate emission of gamma rays or other particles, such as beta particles, alpha particles, fission products, and/or neutrons. This process of creating radioactive materials through the process of neutron absorption is called neutron activation.

1.5.2 Flux Depression

One characteristic of the interaction of a neutron flux with material is the phenomena known as flux depression. It's caused by neutrons interacting with particles as they enter the material, which reduces the number of available neutrons that can cause interactions as they go deeper into the material. The following equation describes the behavior of flux within a given material. Equation 1 displays the fraction of flux that has NOT undergone an interaction at a certain depth, x , in comparison to the original flux as it enters the material.

$$\frac{\phi}{\phi_0} = \exp(-N\sigma x) \quad (1)$$

Where N is the atomic density (atoms/cm³) of the target material, σ is the absorption cross section of the target material, and x is the depth in the material. As can be seen, the percent of neutrons undergoing an interaction within a material decreases exponentially with depth. [5] Therefore, in the scenario where a flux is isotropic on the target (i.e. bombarding from all directions), as is the case in this experiment, this decrease of interactions should cause a lower specific activity in the center of the target.

Chapter 2: Project Objectives

2.1 Project Description

Two sets of six enriched ^{62}Ni foils stacked into layers were irradiated in HFIR to produce ^{63}Ni . Figure 1 shows the orientation of the foils within the aluminum “rabbit” placed in the hydraulic tube of HFIR.

These foils were analyzed in order to determine the difference in specific activity between the inner and outer foils of the sets in order to aid in characterizing the effects of neutron depression on the target. Figure 2 displays a theoretical estimation of the expected fraction of neutron flux remaining in a target that has not yet undergone a neutron absorption interaction if an equal flux were to strike the target perpendicularly from both sides. The data for this plot was acquired by using Equation 1 from the previous section in combination with the cross sections and enrichment concentration of each isotope of nickel in the foils.

Because ^{63}Ni is a purely beta emitting radioisotope, liquid scintillation counting (LSC) had to be used to determine the foils’ activities. Therefore, the foils had to be dissolved in acid before being added to the LSC cocktail. In addition, samples of each foil solution were chemically treated and counted again via liquid scintillation to ensure that no radiocontaminants were negatively affecting the results. These results were then compared to results created through a radioisotope production analytical software called IsoChain and Monte Carlo N-Particle (MCNP) transport code. This project is a continuation of an experiment performed in 2012 by Spencer Walsh who worked with the same group that currently supports this research, the Nuclear and Radiochemistry Group, within the Isotope and Fuel Cycle Technology Division. Her research ultimately proved to be inconclusive, so the design of this experiment comparing stacked nickel foils rather than a sintered pellets and rings, as was used in her experiment, was formulated. [6]

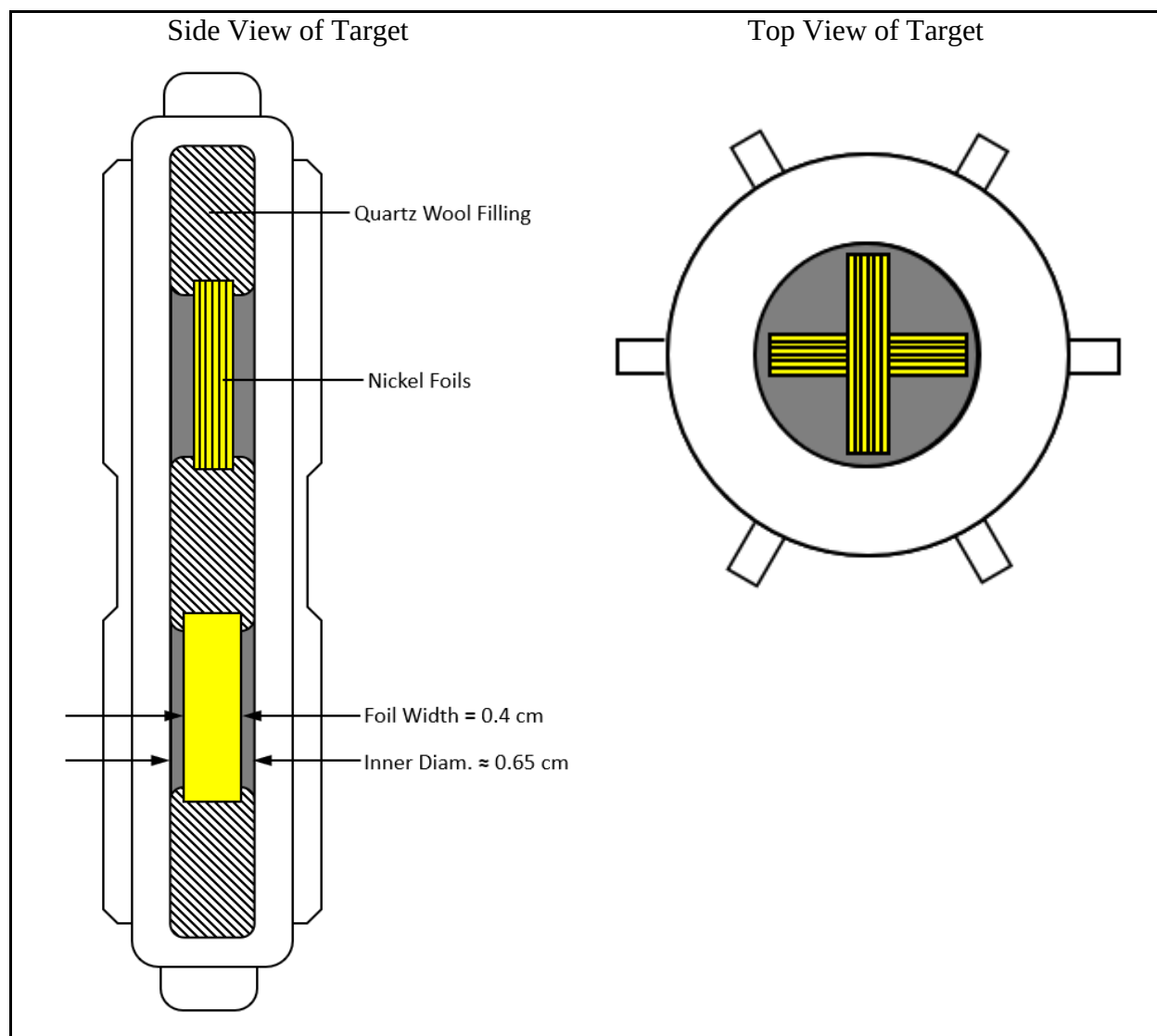


Figure 1: Nickel foil's orientation inside Hydraulic Tube "Rabbit"

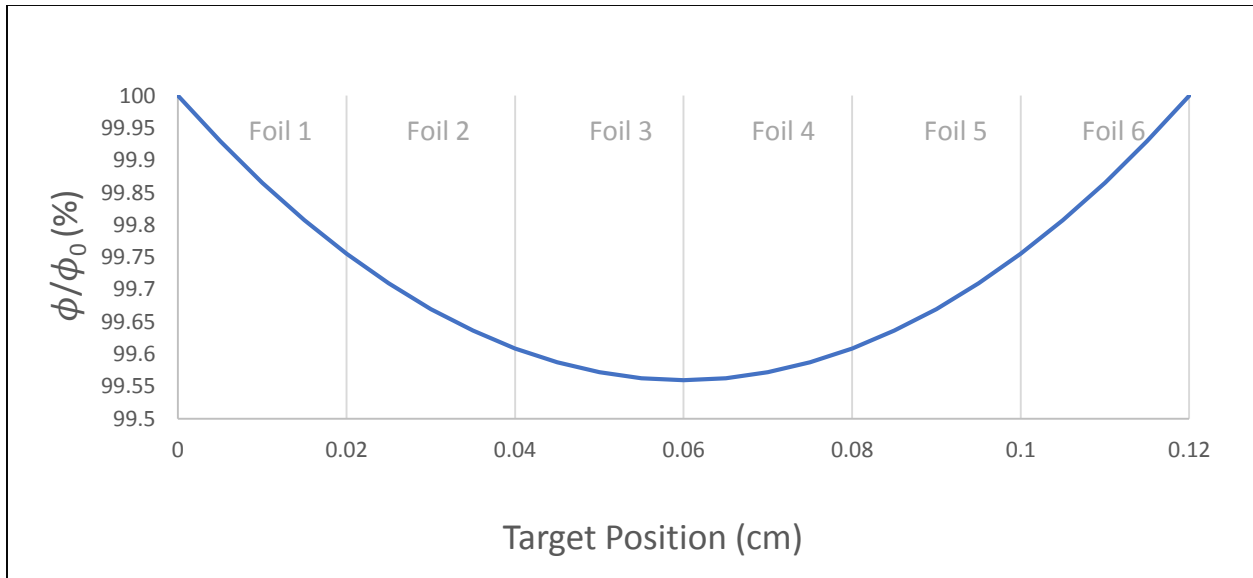


Figure 2: Target flux profile estimation

2.2 Target Description

2.2.1 Foil Composition

The nickel used in the construction of NM-901 target originated from batch 204226 provided by the Isotope and Fuel Cycle Technology Division of ORNL. It was originally created at what is now the Y-12 National Security Complex by a calutron, according to Walsh, but has been stored at ORNL since its production. A preliminary spectrographical analysis of the batch, presented in Figure 3, was taken prior to irradiation. The abundance of the different isotopes of nickel can be found on the left-hand side of the datasheet. The most important feature of this data is the ^{62}Ni enrichment of the samples, which is 86.31%, as it the target to produce ^{63}Ni . The atomic percentage of the isotopes of nickel present in the sample was the foundation for the IsoChain calculation. In addition, all significant impurities listed (>1 ppm) were input into an IsoChain simulatin to get an idea of what radioimpurities might have been produced during irradiation; however, none of the major impurities that the simulation predicted coincided with the impurities seen in the later gamma counts for the samples.

2.2.2 Foil Geometry and Positioning

The nickel was pressed into a thin foil slab in June 2012 with the dimensions of $1.2 \times 4 \times 0.02$ cm^3 , which was then cut into twelve smaller pieces with dimensions of $0.4 \times 1 \times 0.02$ cm^3 . Figure 4 shows the intended cut of the original foil (Note: not to scale). The proportions for the foils were taken directly from documentation created for Walsh [ATTACHMENT 2]. These are particularly important as they are the dimension used for defining the geometries in the MCNP calculation.

These foils were then separated into two stacks of six foils on top of each other and wrapped in aluminum foil to hold them into place. They were then evenly spaced within an aluminum “rabbit”, which is simply a small aluminum alloy container designed for insertion into the hydraulic tubes of HFIR, with the two stacks being oriented perpendicularly to each other. This rabbit was labelled NM-901. The ends and space between the two sets of foil were stuffed

with quartz wool to hold them in place during irradiation. Table 1 displays the irradiation details of the rabbit containing the two stacks of enriched ^{62}Ni in HFIR.

ASSAY																						
Element: Nickel Symbol: Ni Isotope: 62 Series: Batch: 204226	SPECTROGRAPHIC ANALYSIS (SSMS)																					
	Element: ppm	Element: ppm	Element: ppm	Element: ppm																		
ISOTOPIC ANALYSIS <table><tr><th>Isotope</th><th>Atomic Percent</th><th>Precision plus/minus</th></tr><tr><td>58</td><td>8.31</td><td>0.16000</td></tr><tr><td>60</td><td>4.60</td><td>0.07000</td></tr><tr><td>61</td><td>0.48</td><td>0.01000</td></tr><tr><td>62</td><td>86.31</td><td>0.25000</td></tr><tr><td>64</td><td>0.30</td><td>0.01000</td></tr></table> The limits quoted above are an expression of the precision of this measurement only. The error is estimated at less than 1% from known sources of systematic errors. Internal Use Only Date Entered: 02/19/2003 Last Change: 11/26/2003 06:35:12 AM By: MFG	Isotope	Atomic Percent	Precision plus/minus	58	8.31	0.16000	60	4.60	0.07000	61	0.48	0.01000	62	86.31	0.25000	64	0.30	0.01000	Ag: <9.0	I: <.2	Rh: <.4	Tc:
	Isotope	Atomic Percent	Precision plus/minus																			
	58	8.31	0.16000																			
	60	4.60	0.07000																			
	61	0.48	0.01000																			
	62	86.31	0.25000																			
	64	0.30	0.01000																			
	Al: 17.0	In: <.5	Ru: <.4	Te: <.5																		
	As: <.1	Ir: <.4	S: 68.0	Th: <.3																		
	Au: <.8	K: 56.0	Sb: <.3	Ti: 28.0																		
	B: 6.0	Li:	Sc: 2.0	Tl: <.4																		
	Ba: 26.0	Mg: 10.0	Se: <.2	U: <.3																		
	Be: .6	Mn: 25.0	Si: I	V: 82.0																		
	Bi: <.3	Mo: <.5	Sn: <1.0	W: <.8																		
	Br: <.2	N:	Sr: 5.0	Zn: 11.0																		
	C:	Na: 1.0	Ta: <2.0	Zr: 2.0																		
	Ca: 190.0	Nb: <.1	LANTHANIDES and ACTINIDES																			
	Cb: <.1	Ni: M																				
Cd: <.5	O:																					
Cl: 42.0	Os: <.6																					
Co: <.2	P: I																					
Cr: 740.0	Pa:																					
Cs: <.2	Pb: 18.0	Am:	La: 76.0																			
Cu: 47.0	Pd: <.5	Bk:	Lu: .8																			
F: <.3	Pm:	Ce: 71.0	Md:																			
Fe: 27.0	Po:	Cf:	Nd: 82.0																			
Ga: <.5	Pt: 39.0	Cm:	Np:																			
Ge: <4.0	Ra:	Dy: 3.0	Pr: 19.0																			
Hf: <.9	Rb: <.4	Er: <7.0	Pu:																			
Hg: <.9	Re: <.4	Es:	Sm: 3.0																			
		Eu: .6	Tb: .7																			
		Fm:	Y: 40.0																			
		Gd: 5.0	Yb: 2.0																			
		Ho: .7	Tm: <.2																			

* Symbols: M - matrix; S - support; T - trace; I - interference; < - less than; <= less than/equal to; nd - not detected.

* Request No. A-0738; Log # 4875; Notebook No. A-107329-L, pgs. 280 - 283.

* <- No spectrum line visible. Probably absent, definitely less than value given.

* <T- Present but less than value given.

* The spectrographic results reported herein are semi-quantitative estimates and should not be interpreted or construed to be precise quantitative determinations.

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* <- No spectrum line visible. Probably absent, definitely less than value given.

* <T- Present but less than value given.

* The spectrographic results reported herein are semi-quantitative estimates and should not be interpreted or construed to be precise quantitative determinations.

Figure 3: Enriched Nickel Foil Assay

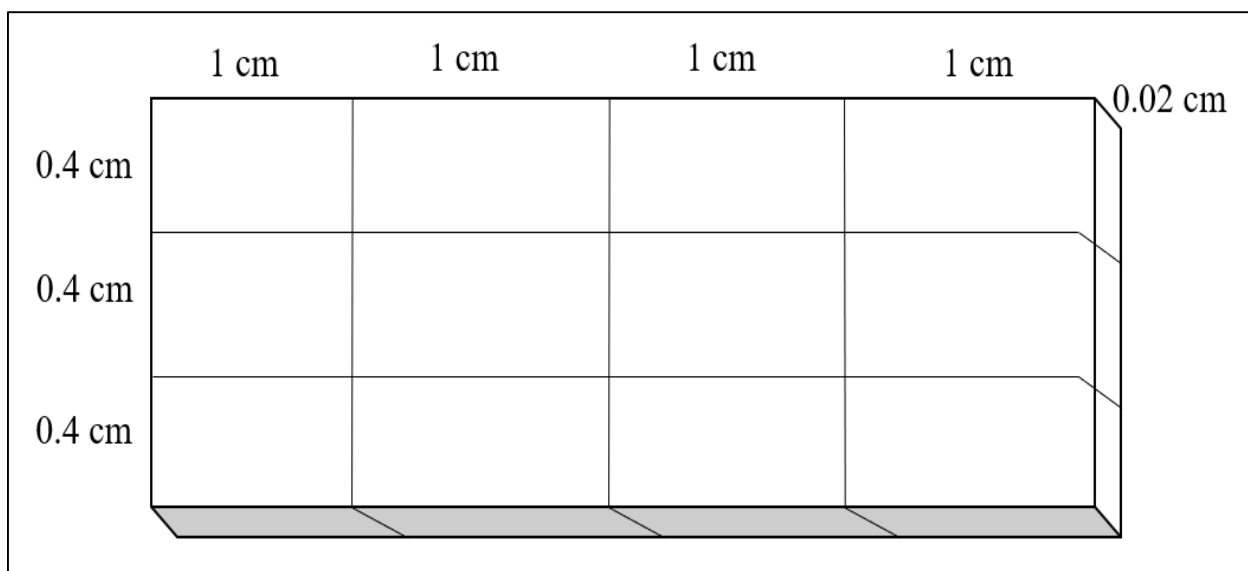


Figure 4: Nickel Foils Dimensions

Table 1: Target NM-901 Rabbit Data

Cycle	443
Location	B3 (The Hydraulic Tube)
Configuration	5
Start Time	8/23/12, 14:44
Finish Time	8/23/12, 16:44
Elapsed Time (hr)	2
Flow	4.8

The data for Table 1 was retrieved from ORNL's EXCITE database [ATTACHMENT 3]

Chapter 3: Theory and Methodology

3.1 Foil Dissolution

Once the foils were removed from the aluminum foil covering, they were placed into individual “counting cards” designed for being placed in the shelving slots of the High Purity Germanium gamma counter in Lab 311. The samples in these cards can be seen in Figure 5. Each foil was counted for 10 minutes to determine radiocontaminants. Next, before placing the foils into glass scintillation vials for dissolution, each foil was removed from their gamma counting card and carefully weighed to determine its mass. They were then placed in individual glass scintillation vials and were moved into Hood IE-8531 of Lab 317A.

Using these weights in combination with the data found in the spectrographical assay (Figure 3), the amount of total nickel as well as the amounts of each isotope of nickel prior to irradiation was calculated and can be found in Table 2 below.

The procedure for dissolving the foils was adapted from the standard procedure for dissolution of commercially produced ^{63}Ni samples created by the NSIT. In this procedure, the ^{63}Ni container is placed on a hot plate set to 200°C and 4.5M HNO_3 is added to their samples. After a short while, a vigorous chemical reaction occurs that results in the rapid dissolution of the sample. However, because the ^{63}Ni produced for commercial applications typically comes in a “chalk-like” state rather than the metal foils created for this experiment, after adding 0.5 mL of 4.5M HNO_3 to the first sample that was attempted to be dissolved, it was deemed that the rate of this chemical reaction would be too slow, taking approximately two hours to dissolve one sample. Therefore, approximately 0.5 mL of 10M HNO_3 was gradually added to the samples with a dropper until the chemical reaction occurred. Take note that due to the imprecision of this method, once the reaction began to occur, the vials were removed from the hot plate in order to slow the reaction, as some would begin to dissolve and bubble rather violently. Figure 6 demonstrates the characteristic green color of the ^{63}Ni samples in solution once dissolved.

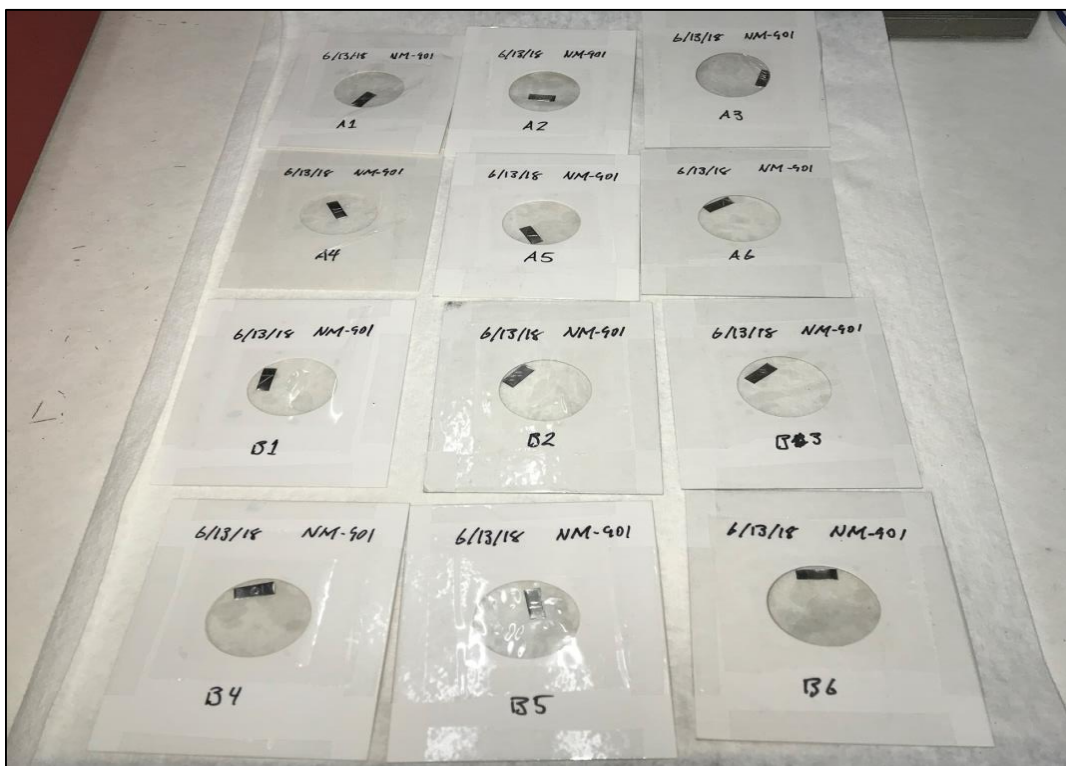


Figure 5: Nickel Foils in HPGe Counting Cards

Table 2: Foil Weights and Isotopic Distribution

	Measured Weight (mg)	Contamination (mg)	Tot Ni (mg)	Ni-58 (mg)	Ni-60 (mg)	Ni-61 (mg)	Ni-62 (mg)	Ni-64 (mg)
A1	82.1	0.15	81.95	6.81	3.77	0.39	70.73	0.25
A2	66.7	0.12	66.58	5.53	3.06	0.32	57.47	0.20
A3	71.5	0.13	71.37	5.93	3.28	0.34	61.60	0.21
A4	72.3	0.13	72.17	6.00	3.32	0.35	62.29	0.22
A5	76.2	0.14	76.06	6.32	3.50	0.37	65.65	0.23
A6	79	0.14	78.86	6.55	3.63	0.38	68.06	0.24
B1	71.1	0.13	70.97	5.90	3.26	0.34	61.26	0.21
B2	81.3	0.14	81.16	6.74	3.73	0.39	70.05	0.24
B3	75.2	0.13	75.07	6.24	3.45	0.36	64.79	0.23
B4	79	0.14	78.86	6.55	3.63	0.38	68.06	0.24
B5	70.9	0.13	70.77	5.88	3.26	0.34	61.08	0.21
B6	67	0.12	66.88	5.56	3.08	0.32	57.72	0.20

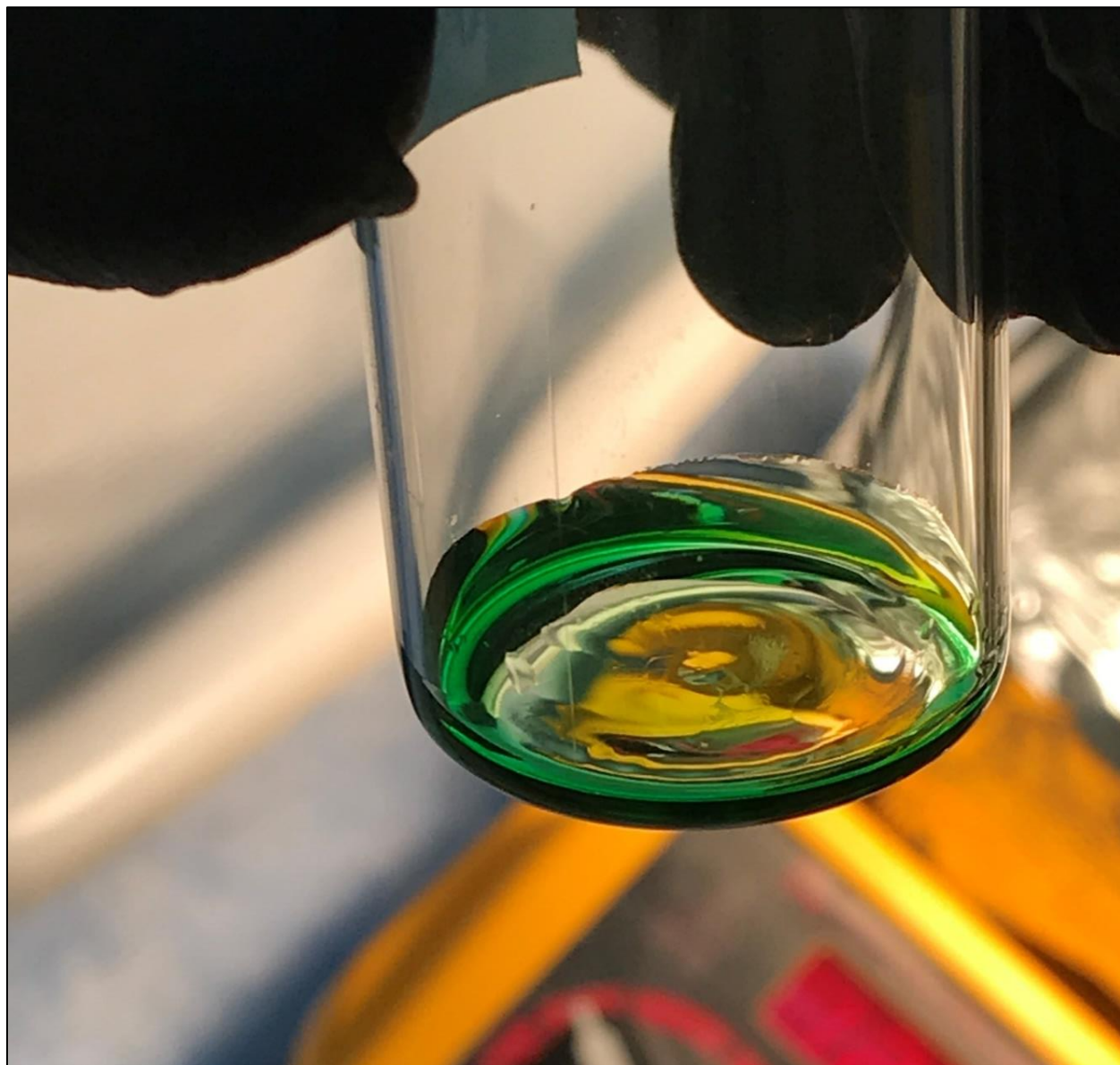


Figure 6: Characteristic Green of Dissolved Nickel Foil

After total dissolution had been reached, each sample then needed to be completely dried. The vials were left on the hot plate set to 120-150°C, a heat lamp connected to a variable autotransformer set to 60% was placed three or four inches directly above the vial, and a tubing/glass pipet system was used to blow a stream of inert gas over the vials in order to expedite the evaporation procedure. Figure 7 shows the typical set up for evaporation in the hood. It was found that once the contents of each vial had been dried to a crystalline state, the lids of the containers had to be securely tightened onto the vial as the samples would return to a liquid state via moisture in the air if the vials were left open.

3.2 Sample Dilution and Liquid Scintillation Counting

As stated previously, because ^{63}Ni is a purely beta emitting radioisotope, the only way to determine the amount of activity present is through liquid scintillation. However, the relationship between the real activity of the sample and the net counts per second determined through liquid scintillation had to be determined by producing an efficiency and quenching curve for the Hidex Triathler Liquid Scintillation Counter in Lab 126 of Building 4501, pictured in Figure 8.

A set of standards was created from a calibrated 5 mL, 5.166 μCi ^{63}Ni standard sample provided by Eckert and Zeigler. This is done by adding 100 μL of the original 5.166 μCi sample into a vial containing 900 μL of 0.1M HNO_3 . Next, 100 μL is then taken from this first dilution and added to another vial containing 900 μL of 0.1M HNO_3 . This process is repeated two more times until four serial dilutions are made. Finally, 100 μL of each dilution is pipetted into 15 mL of Perkin Elmer Ultima Gold scintillation cocktail and are ideally kept in darkness until a series of 5 two-minute measurements are done on each sample as well as a blank sample. A linear regression is then calculated on a Microsoft Excel 2010 file, produced by Justin Griswold of the Nuclear and Radiochemistry Isotope and Fuel Cycle Technology Division, that compares the total counts per second to the actual activity. Figure 9 shows an example of one of these quenching curves, and the slope of the trendline of the standards created is the constant that relates the LSC counts to the sample's actual activity.



Figure 7: Sample Evaporation Setup



Figure 8: Hidex Liquid Scintillation Counter

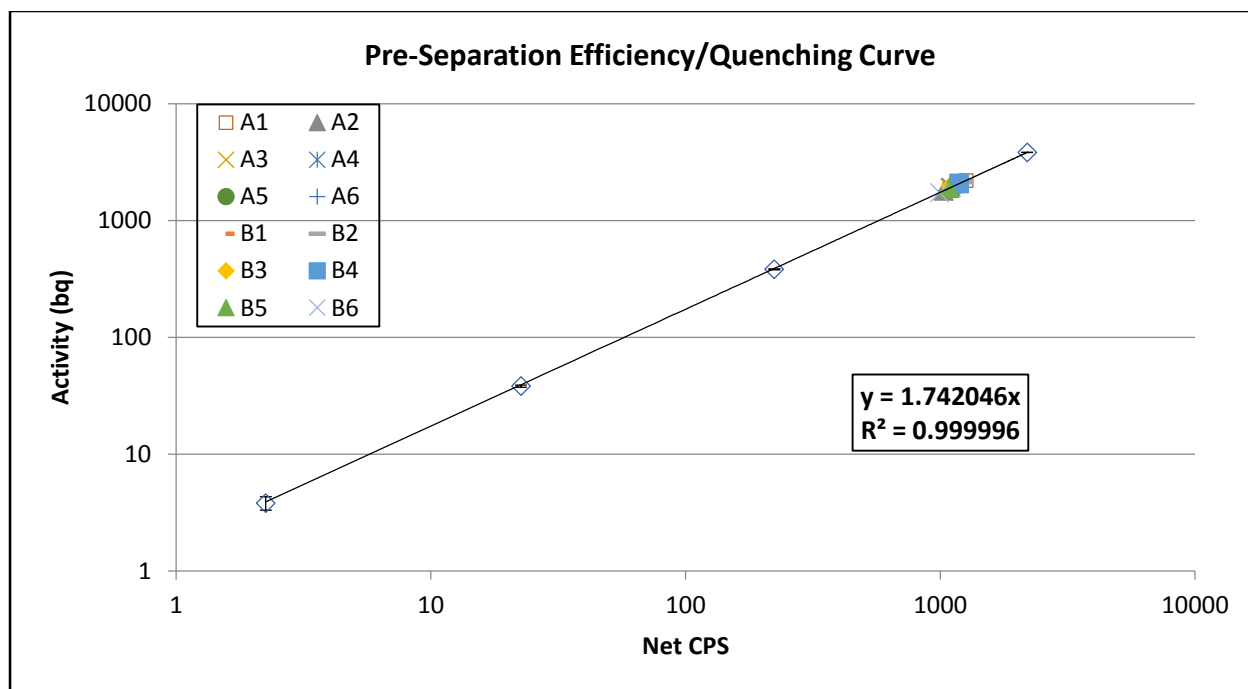


Figure 9: Example Quenching Curve

It should be noted that a new quenching curve was created every time that LSC measurements were done due the fact that cocktail's scintillation ability degrades over time. This caused the relationship between the LSC counts and a sample activity to vary depending on how long the cocktail had been poured; therefore, every time that samples were put into the cocktail for LSC counting, and new batch of standard cocktails were created. This not only ensured accurate immediate measurements, but also allowed for measurements to be taken far after the samples had been made, as both the sample and standard cocktails would degrade at the same rate.

In addition, all samples created had to be diluted in order for them to fit on the quenching curve without extrapolation. Therefore, for the ^{63}Ni samples created in this experiment, once they were dried down, 10 mL of 0.1M HCl was added to each vial, and 100 μL of this solution was taken and added to a different vial containing 9.9 mL of 0.1M HCl, creating an aliquot sample of exactly 10 mL, which were typically labeled "A1-B6 DIL2 / 0.1M HCl". 500 μL were then taken from the aliquot samples, placed into "purple top" transportation vials, which were then placed into secondary containers, empty glass scintillation vials. These were then all placed into a plastic bag and "surveyed out" of Lab 311/317 and transported down to the hood in Lab 126C in Building 4501. Being "surveyed out" is simply the process of checking for any surface contamination on the samples' container via liquid scintillation by the building's radiation control technician. For these initial samples, only one set of LSC cocktails were made for each sample, as opposed to triplicate samples created after chemical separation, by adding 100 μL of the 500 μL transported down into 15 mL of blank LSC cocktail fluid. Like the standard samples, these were all given 5 two-minute counts in the Hidex LSC.

3.3 Experimental Calculations and Error Analysis

The first step in calculating the specific activity of each sample was to calculate the counts for each sample compensated for background. Equation 3 displays the equation for finding the net counts for the sample, C , using the mean of gross counts of the sample, G , and the mean of the background counts, B . Equation 4 displays the appropriate equation for error propagation to calculate the net counts uncertainty, σ_C .

$$C = G - B \quad (2)$$

$$\sigma_C = \sqrt{\sigma_G^2 + \sigma_B^2} \quad (3)$$

Where σ_G is the standard deviation of the five sample counts, and σ_B is the standard deviation of the five background counts, σ_B . Equation 4 below displays the formula for calculating the standard deviation of a set of values, where x_i is the observed count, $\bar{x}$ is the mean value of the counts, and N is the total number of counts.

$$\sigma = \sqrt{\frac{\sum_{i=1}^N (x_i - \bar{x})^2}{N-1}} \quad (4)$$

Next, the specific activity of the whole sample is calculated by converting the counts to count rate, multiplying by the conversion factor attained through the calibration curve (q), (see Section 3.2), multiplying by the dilution factor (DF), converting from Becquerels to millicuries, and, finally, by dividing by the foils original mass, m . This process is displayed in Equation 5:

$$A_{sp} = \left(\frac{C}{120sec} \right) \left(\frac{q \cdot DF}{m} \right) \left(\frac{mCi}{3.7 \times 10^7 Bq} \right) \quad (5)$$

The total relative uncertainty of the specific activity was derived from the following equation:

$$\sigma_{A_{sp}} = \sqrt{\sigma_{RC}^2 + \sigma_m^2 + \sigma_{standard}^2} \quad (6)$$

where

$$\sigma_{RC} = \left(\frac{\sigma_C}{C} \right) \times 100 \quad (7)$$

σ_{RC} is simply the relative uncertainty of the mean counts compensated for background (in percent, σ_m is the uncertainty in the mass measurements for the foils, which was assumed to be 1%, and $\sigma_{Standard}$ is the error inherent with the standards, a value determined by documentation for the original standard sample from Eckert and Zeigler [ATTACHMENT 4].

3.4 Liquid Chromatography

After an initial activity analysis for each sample was completed via the procedure described above, it was decided that it was possible that the data may be altered by other beta emitting particles in the solution, namely, the two radiocontaminants determined through the gamma counts, Co-60 and Eu-152. A procedure was then developed to chemically separate out these contaminants, which was executed in the same hood as the dissolution and dilution procedures.

3.4.1 Column Construction

Prior to the actual separation procedure, the most appropriate separation column for the two radiocontaminants had to be determined and produced for each sample. These were made in the dry lab in 4501, Lab 208, to reduce possibility of contamination while in the primary lab of this experiment, Lab 317A. The first column for separating out Co-60 from each sample was based off the columns used for the same purpose during the commercial production of ^{63}Ni . Anion exchange columns using 0.4 mL of MP1 resin with a mesh size of 200-400 micron were created and washed with five bed volumes (2 mL) of 10M hydrochloric acid as the solvent for the sample. Unfortunately, the quantity of Eu-152 seen during commercial production never surpassed the allowable radiocontaminant requirements for the purchaser; therefore, there was no standardized procedure for its separation. After a few trial and error attempts, it was determined that a cation exchange column using 0.4 mL of AG50x8 (the “x8” denotes the cross-linking factor) with a mesh size of 200-400 microns washed with five bed volumes (2 mL) of 1M HCl was an effective method for separating the Eu impurities from the Ni. It should also be noted that the columns were stored prior to use by washing them with 5 mL of deionized water after their acid washes and capping them with a small amount of water still in the column. Directly before

use, the columns were pre-equilibrated by washing 2 mL of 10M into the MP1 columns and 2 mL of 1M HCl into the AG50x8 columns into a vial dedicated solely for column conditioning.

3.4.2 Co-60 Removal (MP1 Column)

The procedure for ^{60}Co (and ^{152}Eu in the section below) removal will be described for foil A1; however, this procedure was used for all the samples. Firstly, 2 mL of the original “NM901 A1” sample solution was pipetted into a new glass scintillation vial, labeled “A1 SEP1 / 10M HCl”, and dried using the drying setup described previously. The vial was then closed and bagged out of the hood and taken to Lab 311 for a 15-minute gamma count. After the count, the vial was returned to the hood where 5 drops of 10M HCl were added and it was again dried down in order to ensure that all nitrogen molecules have been removed. Next, 0.5 mL of 10M HCl was added to the vial, which was then placed on a hot plate set to 50°C to expedite the dissolution of the ^{63}Ni material. Once the solution was completely translucent, a glass pipet dedicated solely to this sample and this step in the procedure was used to suck out the solution and add it to the MP1 column. The solution that passed through was collected in a separate vial labeled “A1 MP1Cl / L1”. Once the liquid had passed through the column, the “SEP1” vial was then rinsed with another 0.5 mL of 10M HCl, which was again sucked out using the glass pipet and added to the MP1 Column. This was repeated two more times, allowing a total of 2 mL to pass into the “A1 MP1Cl / L1” vial. The glass pipet for this step was then discarded into a “Sharps/Tips” waste container after this step. The “A1 MP1Cl / L1” vial was then replaced with a vial labeled “A1 MP1Cl / W1”, and the column was washed directly with 2 mL of 4M HCl. Finally, the “A1 MP1Cl / W1” vial was replaced with a “A1 MP1Cl / E1” vial, and the column was washed with three bed volumes (1.2 mL) of deionized water. Also, during the time it took to complete the last two washes, the “A1 MP1Cl / L1” vial was made to undergo the evaporation procedure described previously, and once completely dry, it was bagged out from the hood and given another 15-minute gamma count.

Figure 10 shows the MP1 column in use during the first wash. Figure 10a depicts the dried down contents of the original 2 mL of the A6 sample. Figure 10b shows the sample after being fully dissolved in 10M HCl. Figure 10c depicts the MP1 column as the contents of the A6 load is initially added to it. Figure 10d displays the column after the initial wash has passed through and the second rinse of 0.5 mL of 10M HCl has been added.

3.4.3 *Eu-152 Removal (AG50x8 Column)*

For this column, the “A1 MP1Cl / L1” vial was treated very similarly to the “A1 SEP1 / 10M HCl” vial in the previous column; however, there are quite a few easily confusing variations in this procedure, as will be discussed. The contents of the “A1 MP1Cl / L1” was completely dissolved using 0.5 mL of 1M HCl, and a new glass pipet was used to suck out the liquid and add it to the AG50x8 column. The solution that passed through was collected in a separate vial labeled “A1 AG50Cl / L1”. The “A1 MP1Cl / L1” vial was then rinsed with 1 mL of 1M HCl, which was then sucked out and added to the column after the contents of the previous wash had completely passed through. This step was then repeated for two more times, allowing a total of 3.5 mL to pass into the “A1 AG50Cl / L1” vial, which was then replaced with an “A1 AG50Cl / W1” vial. The glass pipet was again discarded at this point. For this vial, 1 mL of 1M HCl was added directly into the column to ensure that the Eu-152 trapped in the resin had not migrated too far down the column and was possibly passing through. These two vials, “A1 AG50Cl / L1” and “A1 AG50Cl / W1” were then bagged out and given individual 15-minute gamma counts. Also, the “A1 AG50Cl / W1” vial was replaced by a “A1 AG50Cl / E1” vial, and the column was washed with 2 mL of 8M HNO₃, which removed the majority of the Eu-152 from the resin. Finally, the “A1 AG50Cl / E1” vial was replaced by a “A1 AG50Cl / WW” vial, and the column was washed with three bed volumes (1.2 mL) of deionized water.

Figure 11 shows the AG50x8 column in use. Figure 11a is the dried down A6 MP1Cl / L1” sample after taking a 15-minute gamma count. Figure 11b is the same vial after dissolving the sample in 1M HCl acid. Figure 11c was taken right after adding the solution was added to the column. Figure 11d shows how the green of the sample slowly moves through the column after a single wash.

a.



b.



c.

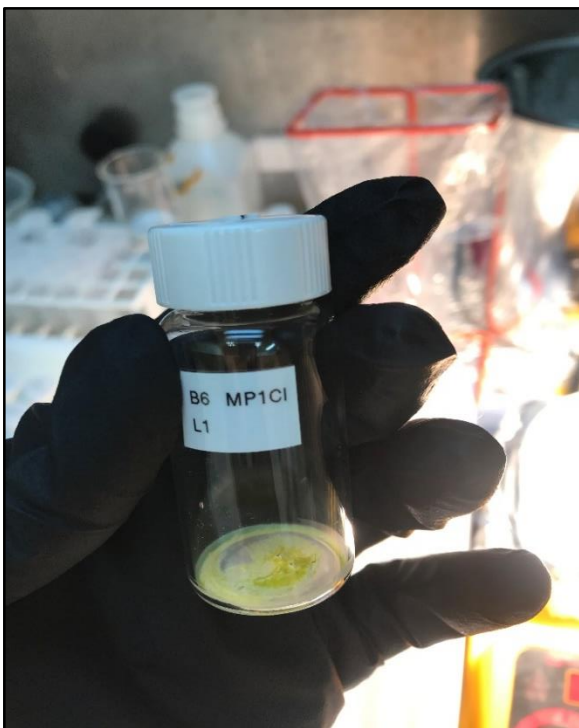


d.



Figure 10: MP1 Column

a.



b.



c.



d.



Figure 11: AG50x8 Column

3.4.4 Post-Separation Processing

Once the gamma counts on “A1 AG50Cl / L1” and “A1 AG50Cl / W1” verified the removal of all Co-60 and Eu-152, the two were placed back into the hood in 317A, and a new glass pipet was used to suck the content of “A1 AG50Cl / W1” and add it to “A1 AG50Cl / L1”. The “A1 AG50Cl / W1” vial was then rinsed twice with 1 mL of 1M HCl, giving a total of 6.5 mL of solution in the “A1 AG50Cl / L1”, which was then relabeled “A1 AG50Cl / L1+W1”. The “A1 AG50Cl / L1+W1” vial was then placed in the evaporation setup and allowed to completely dry down, which was typically done throughout the day in conjunction with the chemical separation of the next sample. Once dried, the sample was bagged out and given a final gamma count for 15 to 36 hours, depending on time availability. Figure 12 displays the positioning of the samples within the HPGe counter.

Figure 13 displays three gamma spectra of the most importance for each sample. Figure 13a displays the initial gamma spectrum for the dried 2 mL of each sample taken before any chemical separation. Figure 13b shows the gamma spectrum for a sample after going through the MP1 column (note the loss of the two Co-60 gamma peaks). Figure 13c displays the gamma spectrum for the sample after going through the AG50x8 (and MP1 prior) column, showing virtually no gamma peaks after the removal of Eu-152.

After the final count, the sample was returned back to the hood and 2 mL of 0.1M HCl was used to dissolve the material. A vial labeled “A1 DIL2 PS / 0.1M HCl” was then filled with 9.9 mL of 0.1M HCl, and 100 μ L from the “A1 AG50Cl / L1+W1” vial was added to it, creating an aliquot sample of exactly 10 mL. 500 μ L was then taken from this vial and put into a purple-top transportation vial, which, along with the rest of the sample transportation vials, was smeared out of Lab 311/317 and taken down to 126A. From here, to create better statistical accuracy, three identical LSC cocktail samples were made for each sample by adding 100 μ L to 15 mL of the cocktail fluid, using up a total of 300 μ L for each of the 500 μ L samples transported. Every sample was also given 5 two-minute counts in the Hidex LSC. A simplified diagram of the chemical separation procedure starting at the “SEP1” vials up until the “purple top” transportation vials is displayed in Figure 14.



Figure 12: HPGe Setup

a.

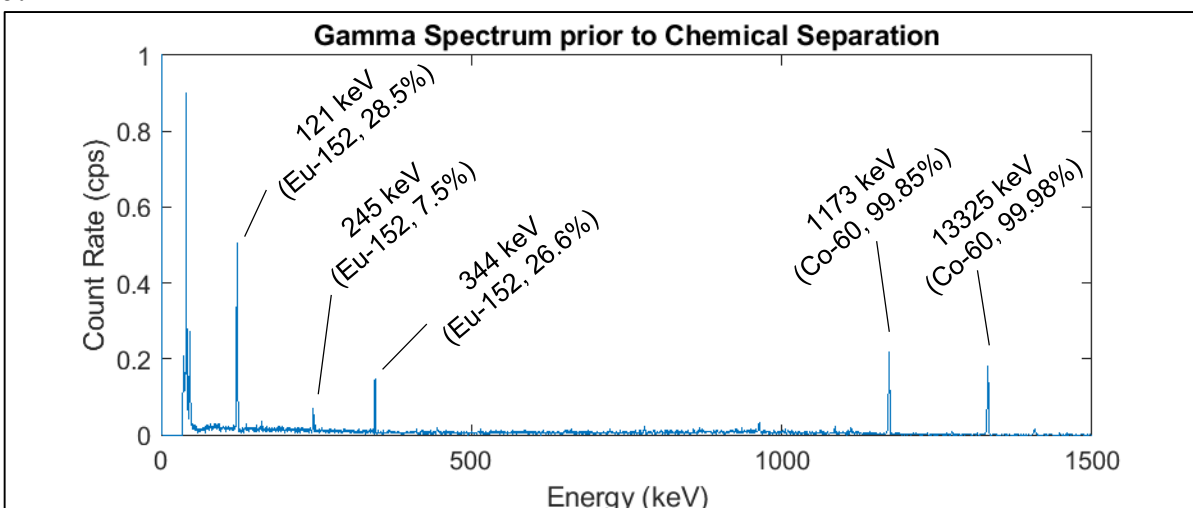
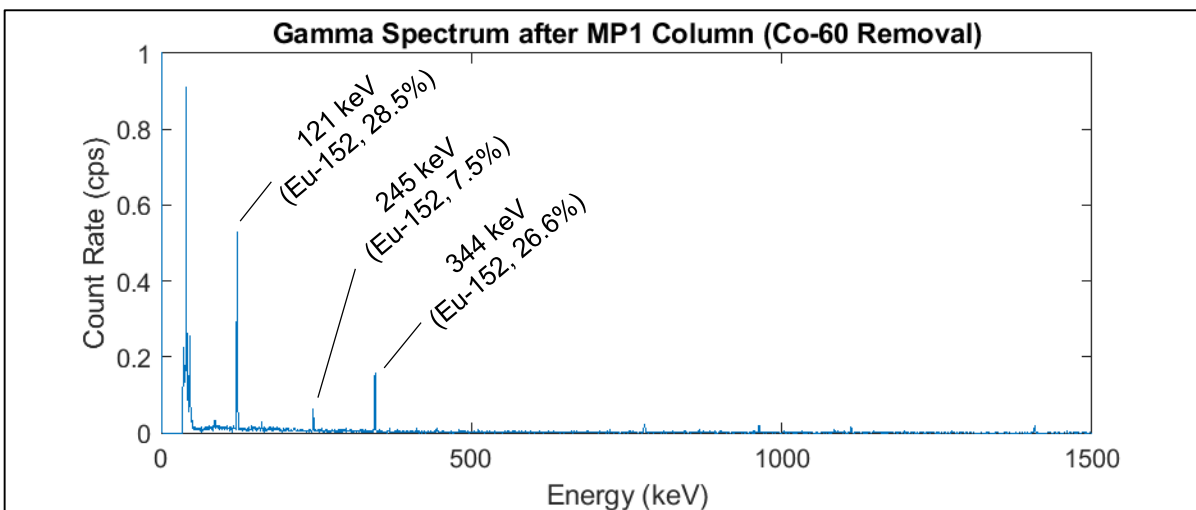


Figure 13: Gamma Spectra during Chemical Separation

b.



c.

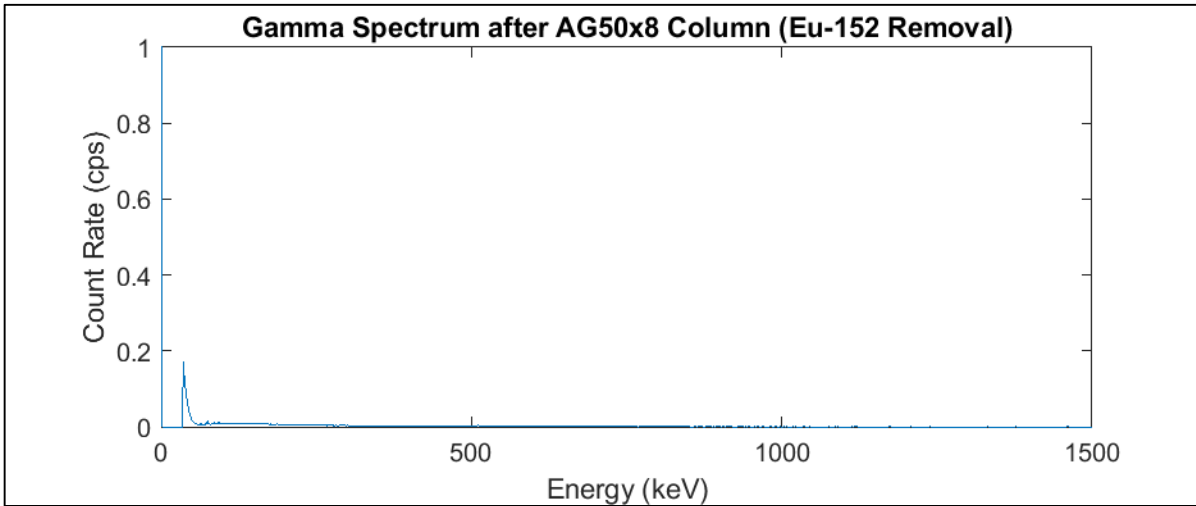


Figure 13 (continued)

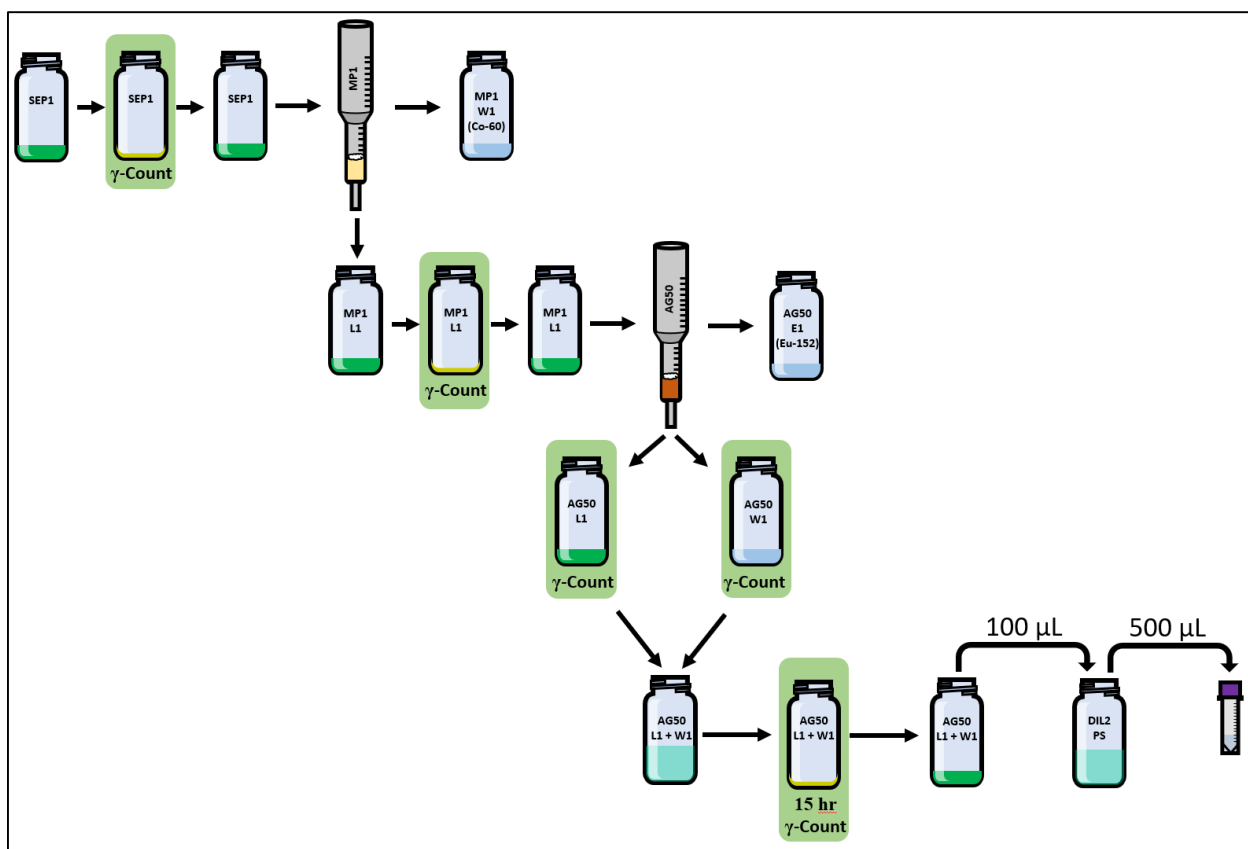


Figure 14: Chemical Separation Diagram

3.4.5 Error Corrections

Correction counts were done for a few of the pre-separation and post-separation samples due to results not corresponding to the expected trends. For some samples, their specific activity was simply unreasonably high or low in comparison to the other samples. For other measurements, the post-separations were higher than those for pre-separation, indicating unquestionable invalidity. It is suspected that variation in the volume of 0.1M HCl during dilution of the samples is the primary cause for the unreliable data. Therefore, new dilutions for the samples were done, and they were recounted. The resulting data for these corrections were averaged with the original values collected. The pre-separation samples that were reevaluated are A2, B5, and B6. The post-separation samples that were reevaluated include A1, A3, and B6.

It should also be noted that sample A1 experienced slight setbacks during the chemical separation process. Because it was the first sample treated, the percentage of material lost was significantly greater than the other foils. It ran through an extra AG50x4 column, which didn't work as expected and prompted the use of the AG50x8 column.

3.5 IsoChain Theory

Isochain is a Java-based analytical tool used to predict nuclide production and decay in a given constant neutron flux. It was created as an alternative to programs that utilize classic Bateman equation (Equation 8) in order to overcome issues that arise when trying to solve for transmutation and decay problems with equal depletion constants. This equation displays the number of atoms, $N_n(t)$, of the n^{th} species at time t , where N_k^0 is the number of atoms of species k at time $t=0$ and c_i is the formation rate constant (λ_n or $\sigma_n\phi$).

$$N_n(t) = \sum_{k=1}^n (\prod_{i=k}^{n-1} c_i) N_k^0 \sum_{i=k}^n a_i e^{-\Lambda_i t} \quad (8)$$

where

$$a_i = \prod_{j=i}^{n-1} (\Lambda_j - \Lambda_i)^{-1} \quad (j=k, k+1, \dots, n) \quad (9)$$

and

$$\Lambda_n = \lambda_n + \sigma_n\phi \quad (10)$$

In Equation 10, the term Λ_n is the total depletion constant, λ_n is the decay constant (s^{-1}) of nuclide n , σ_n is the neutron capture cross section (cm^2) on nuclide n , ϕ is the neutron flux ($n\ cm^{-2}\ s^{-1}$). However, IsoChain can overcome this through numerical calculations that recursively integrate the transmutation and decay chains into separate linear calculations, as displayed in Equation 11.

$$N_n(t) = N_1^0 (\prod_{i=1}^{n-1} c_i) D_n(\Lambda_1, \dots, \Lambda_n)(t) \quad (11)$$

where

$$D_n(\Lambda_1, \dots, \Lambda_n)(t) = e^{-\Lambda_n t} \int_0^t D_{n-1}(t') e^{-\Lambda_n t'} dt' \quad (12)$$

The variable solved by Equation 12, $D_n(\Lambda_1, \dots, \Lambda_n)(t)$, represents a depletion factor for nuclide n , which is what is recursively integrated during each subsequent depletion factor. [7]

For this this experiment, 1 gram of nickel with isotopic proportions that of the enrichment specified by the spectrographic assay of the foils (Figure 2) was bombarded by the following flux specifications found in Table 3.

One of the downfalls of this program, however, is the lack of an up-to-date library of isotope cross sections. Therefore, the cross sections for many of the isotopes had to be manually adjusted and were set to experimentally determined values collected and released by Mughabghab [ATTACHMENT 1]. Table 4 presents the absorption cross section and resonance integral data used in the theoretical ^{63}Ni production calculations.

In addition, IsoChain allows for users to input a range of isotopes that the calculations consider. Figure 15 depicts the input screen for IsoChain after the range of consideration has been set; therefore, only the isotopes included in this figure were accounted for during its analysis. It should be noted that the values for IsoChain are calculated considering only the amount of nickel present, the neutron flux specified, and the amount of time it is bombarded. The model does not compensate for flux depletion of any kind

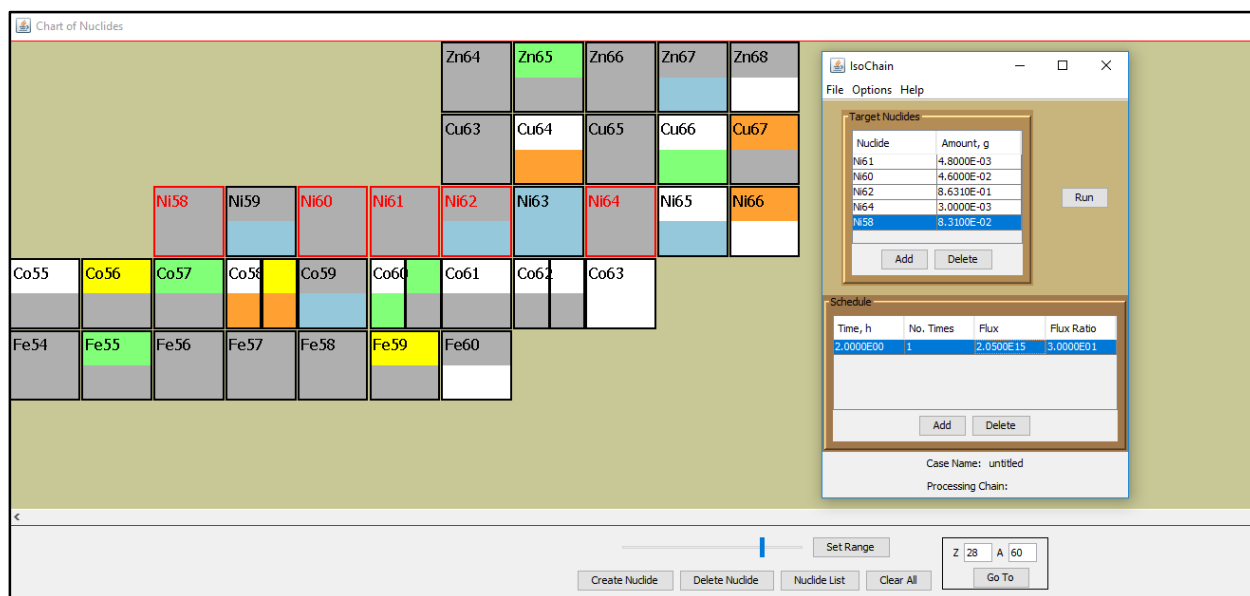
Table 3: IsoChain Case Specifications

Flux (ϕ_0)	2.05E15
Flux Ratio (ϕ_0/ϕ_{ep})	30
Time (hr)	2

Table 3 determined the flux and flux ratio values from Reference [8].

Table 4: IsoChain Cross-Section Data

ENDF Material	Source	Capture σ (b)	Capture RI (b)
26-Fe- 54	Mughabghab 2006	2.25	0.18
26-Fe- 56	Mughabghab 2006	2.59	0.14
26-Fe- 57	Mughabghab 2006	2.48	0.3
26-Fe- 58	Mughabghab 2006	1.32	0.03
27-Co- 58	Mughabghab 2006	1900	200
27-Co- 58M	Mughabghab 2006	140000	10000
27-Co- 59	Mughabghab 2006	37.18	0.06
28-Ni- 58	Mughabghab 2006	4.37	0.1
28-Ni- 59	Mughabghab 2006	77.7	4.1
28-Ni- 60	Mughabghab 2006	2.5	0.006
28-Ni- 61	Mughabghab 2006	2.1	0.4
28-Ni- 62	Mughabghab 2006	14.9	0.3
28-Ni- 64	Mughabghab 2006	1.64	0.04
29-Cu- 63	Mughabghab 2006	4.5	0.02
29-Cu- 65	Mughabghab 2006	2.17	0.03

**Figure 15:** IsoChain Input Screen

3.6 MCNP6.2 Theory

MCNP is a general-purpose Monte Carlo N-Particle code used to simulate the random nature of nuclear radiation. It uses Monte Carlo statistics as a foundation for its continuous-energy, time-dependent, radiation-transport code. It is export-controlled and distributed by the Radiation Safety Information Computational Center (RSICC) [website](#). US citizens can request either the distribution package, which includes source code, or the executable only distribution package, and non-US citizens can request the executable only distribution package.

3.6.1 MCNP Structure

To get a better understanding of the simulation created for this particular experiment, one should first understand the general layout of MCNP code structure. MCNP code is broken up into three primary blocks, which also contain subblocks that can alter and further specify the desired code function. The three primary cards and their basic functions are as follows (NOTE: the term *card* is used simply to denote a specific line of code):

- Cell Cards - define the boundaries of material within geometries specified in the surface cards, what material occupies that volume, and the importance of that material in regard to particle interaction
- Surface Cards – defines the geometries of the desired objects within the program
- Data Cards – defines the following parameters (each is its own type of card/subblock):
 - Materials compositions
 - Radioactive Source Specifications (e.i. position, geometry, energy, particle emission type)
 - Particle Physics Mode (e.g. the particle of interest for the simulation)
 - Cell and Surface Parameters (e.g. which cells/surface to track particles within)
 - Tally Specifications (e.g. what the program records – e.i. surface current, surface flux, track length estimate of cell flux, etc.)
 - Problem Termination (e.g. how long to run program or how many particles to simulate)

The order in which these cards are listed are the required order of input for MCNP code, apart from the order that the various data cards are placed within that block. For further specifications on the formatting and use of these cards, see Reference [9].

3.6.2 Experiment Specifications

The HFIR MCNP model of Cycle 400, created by Ned Xoubi and Trent Primm [10] was altered to reflect the scenario of this experiment. Although it was not the same cycle and the alteration of material within the flux trap of HFIR does change the flux slightly, there was no model created for Cycle 443, and to correct all the materials within the core was outside the scope of this thesis. The altered model input code used for this thesis is given in [ATTACHMENT 5].

The first alteration of the code was adding in Surface Cards to create the geometry of the foils, their aluminum casing, and the aluminum rabbit. As stated before, the geometry for the individual foils is based on the foil construction documentation, which is demonstrated in Figure 3, and the positioning of the rabbit containing the foils was determined using the specifications found with the EABD Database, presented in Table 1. Within the model these surface cards include 9466 through 9482. Next, cell cards were created for these geometries. These are cell cards 765, 764, and 9767 through 9780. For the data cards, the first alteration made was the addition of a new material, m999 in the input code, and was created to accurately reflect the isotopic makeup of the enriched nickel foils, which can be found in Figure 2. One thing to note about the material card assigned to the foils in the simulation is that none of the contaminants were included, as the foil was purified to be 99.8% pure nickel. This was a choice of convenience as no one element reached even a tenth of a percent of the compositional makeup of the foil. Also, in order to input this tiny amount of each element into MCNP, the element would also have to be further broken down into the natural abundance of each isotope of the element. The particle mode for tally purposes and cell/surface parameters were set to neutrons and photons, and the program termination input was set to stop when the uncertainty for all the tallies reached less than 0.05%. The source specifications were left as they were. Lastly, track length estimates of cell flux tallies, F8 tallies, were performed for the cells representing the nickel foils. These are simply measurements of the estimated thermal neutron fluxes through each foil.

Energy bins of $2.5\text{E-}8$, $1\text{E-}6$, 0.1 , and 20 MeV were specified for the tallies, although only the data for thermal ($2.5\text{E-}8$ MeV) to slow ($1\text{E-}6$ MeV) neutrons relevant for this work and is reported in the results. It is important to note that the number of particles simulated in this measurement is arbitrary, as it would be beyond the scope of this project to accurately replicate the complex environment of HFIR in regard to the flux the target received within the two hours it was placed in the hydraulic tube. The code was simply allowed to run until the uncertainty of the results were below 1%.

Figure 16 displays various aspects of the geometries of the simulation using a built-in 3D modeling functionality of MCNP called Interactive Plotter. Figure 16a shows the flux trap portion of HFIR from above, and the hydraulic tube can be seen in the center left as the only circle with a pink center. Figure 16b shows the position within the hydraulic tube that the rabbit for this experiment was placed during irradiation as well as the foils within the rabbit. Figure 16c is simply a zoomed in version of Figure 16b that shows the individual foils within one set and how the two set are positioned perpendicularly to each other.

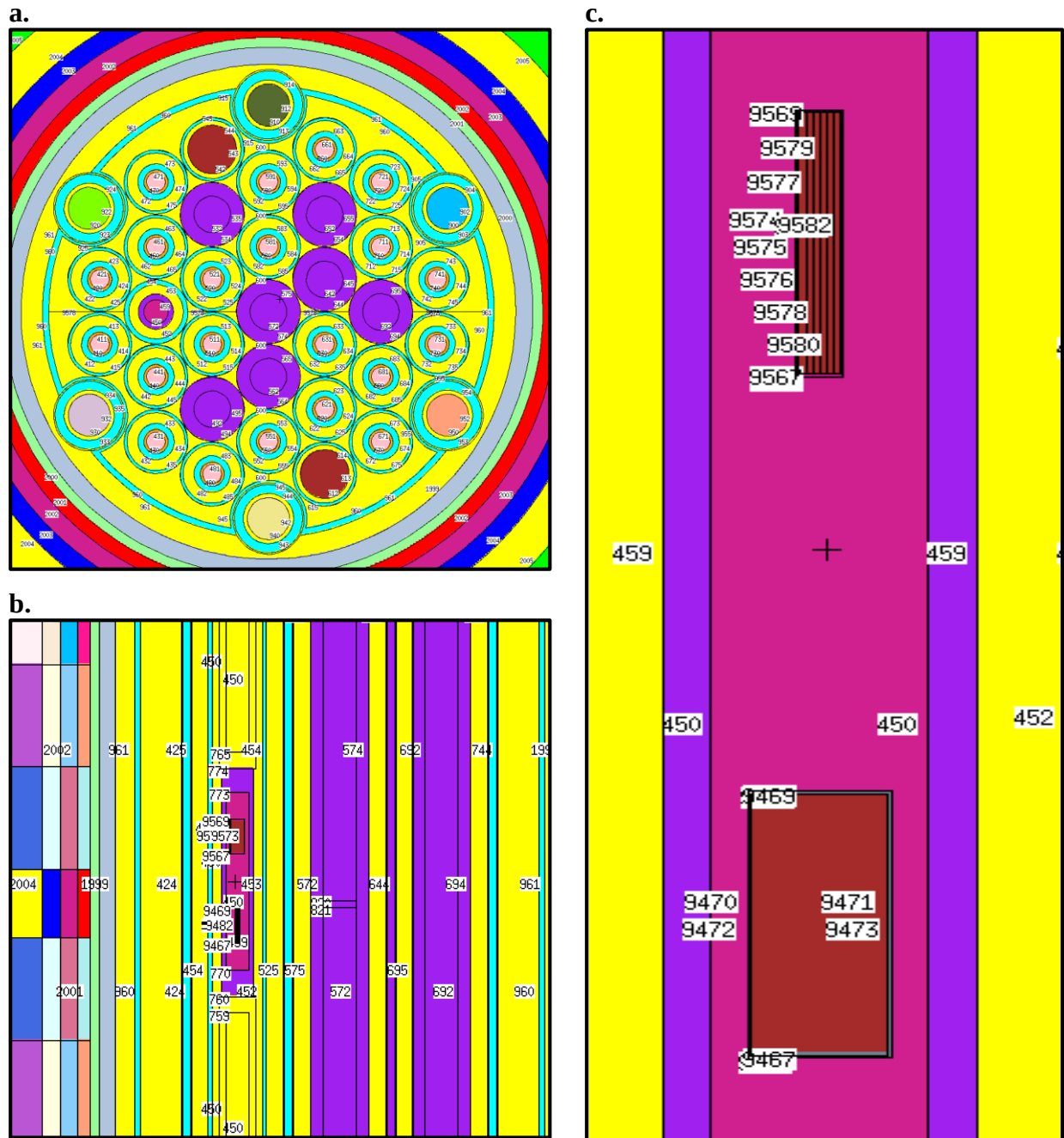


Figure 16: MCNP Geometries Snapshots

Chapter 4: Results

4.1 Pre-Separation Results

Figure 17 and Figure 18 show the specific activity in relationship to the foil positioning for samples A1 through A6 and B1 through B6, respectively. The initial results for the specific activity of the foils were rather surprising, showing very little fluctuation in their values. The increase in specific activity samples A5 and A6 are the only substantial values supporting the effects of flux depression for set A; however, the overall shape of set B is much more align with the expected trends. Regardless, the uncertainties inherent with the measurements overlap with all values for the foils, which casts doubt on to the validity of these trends.

4.2 Post Separation Results

Again, Figure 19 and Figure 20 show the specific activity in relationship to the foil positioning for samples A1 through A6 and B1 through B6, respectively, after chemical separation had been performed. For these plots, the average of the three triplicate measurements for each foil was presented.

A slight trend of neutron depression can be noticed in these results (higher in the outer foils and lower in the inner foils); however, the scale of the plot makes it more prominent. With a larger y-axis scale, these plots look practically linear. The greatest difference between foils for each set is only 6.5% (A1 to A6) and 5.3% (B3 to B6). However, because sample A1 experienced setbacks during chemical separation, as mentioned in Section 3.4.5, the comparison between A1 and A6 should be discounted. The foil with the second lowest specific activity for that set, A3, shows a 6.3% decrease compared to A6, supporting the hypothesis of neutron depression.

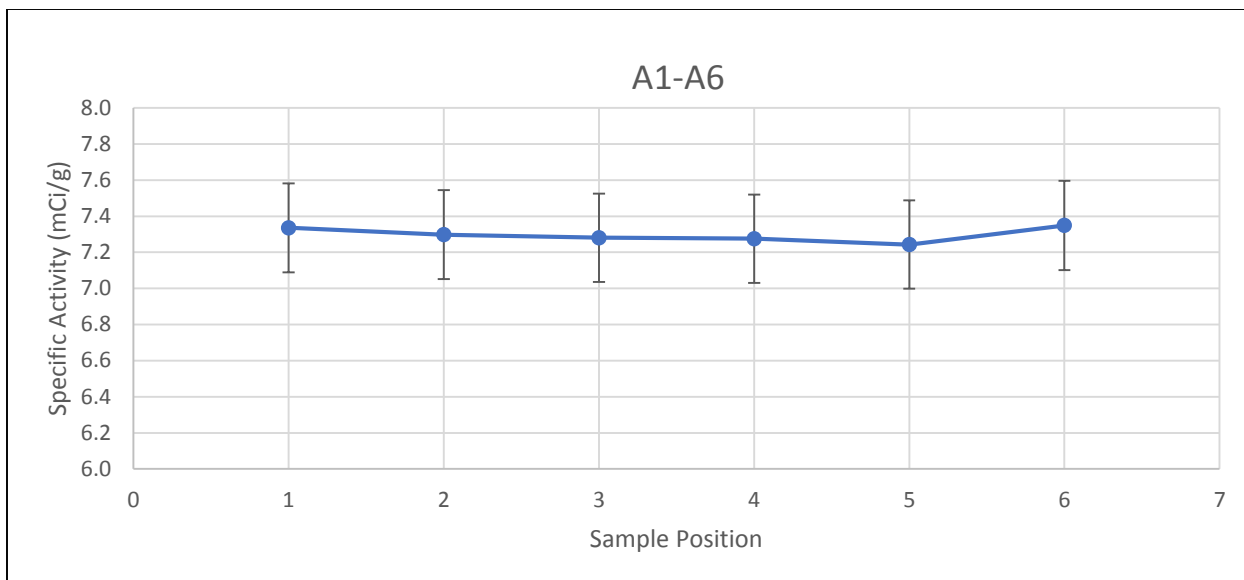


Figure 17: Specific Activity of Samples A1-A6 before Chemical Separation

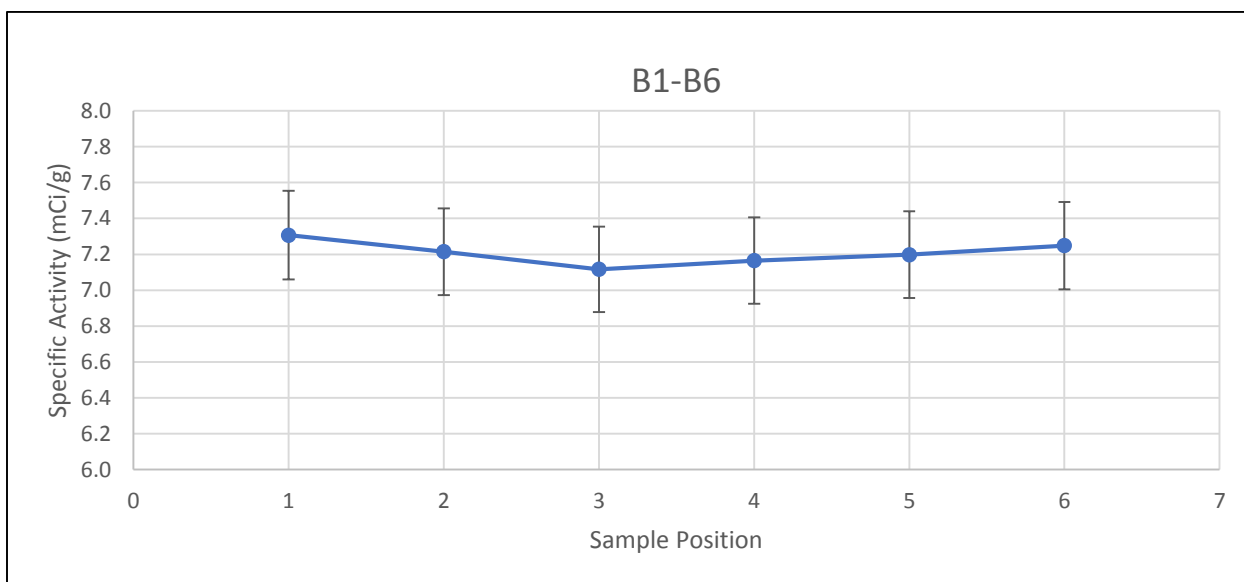


Figure 18: Specific Activity of Samples B1-B6 before Chemical Separation

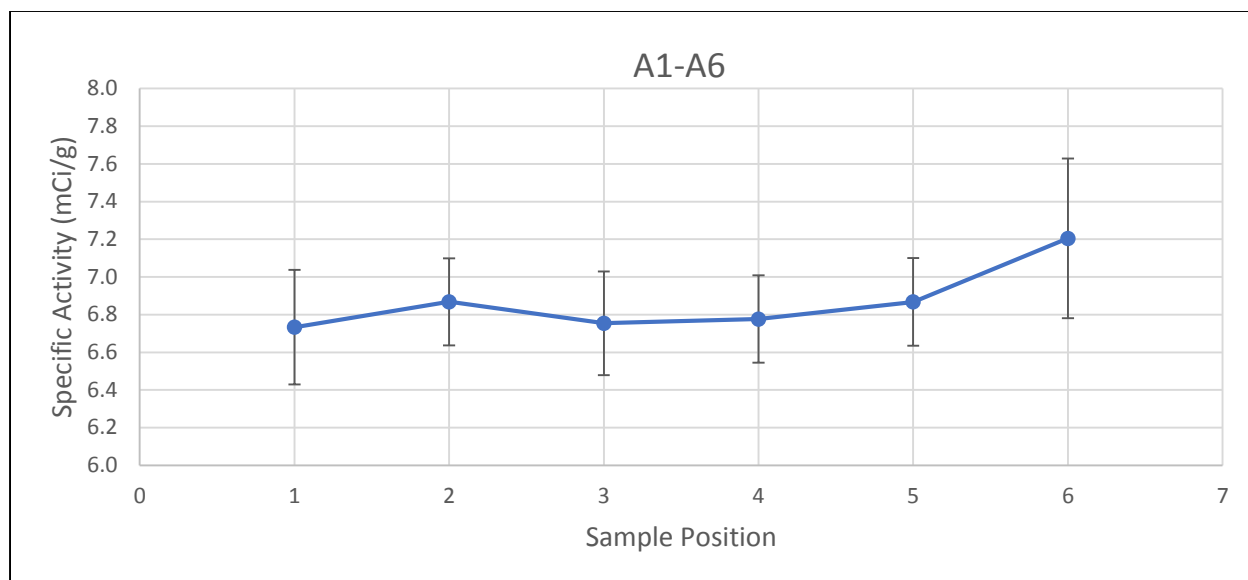


Figure 19: Specific Activity of Samples A1-A6 after Chemical Separation

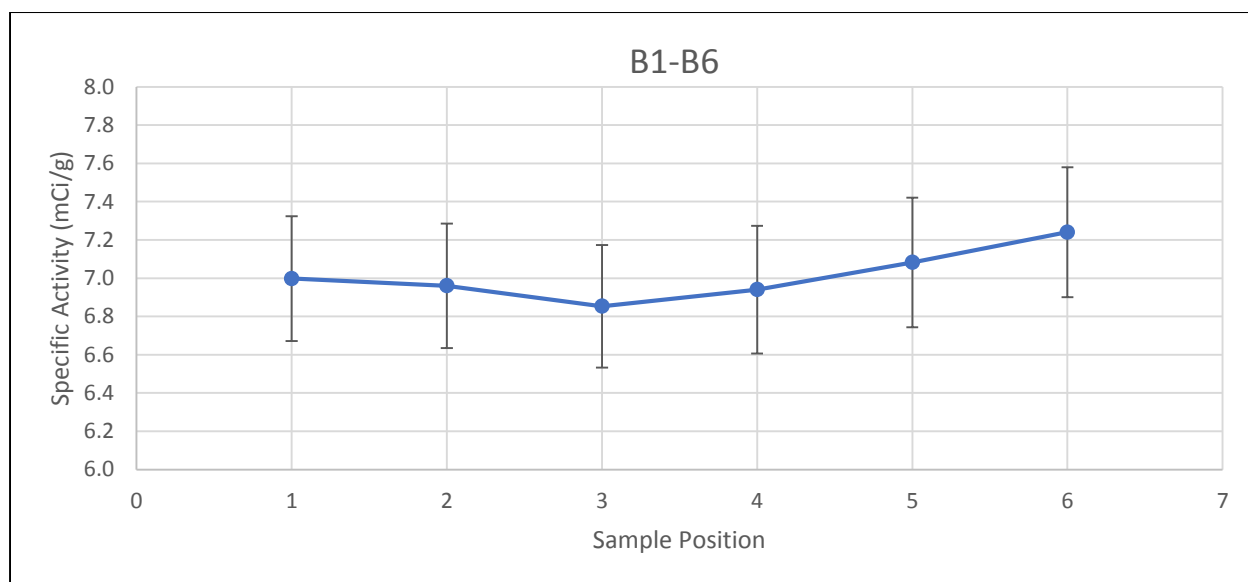


Figure 20: Specific Activity of Samples B1-B6 after Chemical Separation

Table 5 highlights the change in specific activity between the pre-separation measurements and post-separation measurements. The average of the three triplicate samples for each foil during the post-separation measurements was used for these comparisons. It is difficult to know how much of the activity lost was due to the removal of Co-60 and Eu-152 and how much was ^{63}Ni residue left on the containers and columns during the chemical processing.

Also, because the neutron flux should strike both sets of foils isotropically, an average of specific activity was taken between the two sets based on foil position (e.g. A1 and B1), which is presented in Figure 21. Again, the slight U-shaped trend of higher specific activity on the outside and lower on the inside can be noticed. For the averages, there is only a 1.7% variation in the lowest (Position 3) and highest (Position 1) foils for the pre-separation data, and there is a 5.8% variation in the lowest (Position 3) and highest (Position 6) foils for the post-separation data. However, like the other results, the error margins for these measurements are significantly higher than the differences between them.

Table 5: Post-Separation vs. Pre-Separation Comparison

Target Set	Target Position	Post-Sep (mCi/g Ni)	Pre-Sep (mCi/g Ni)	% Difference
A	1	6.73	7.34	8.22
	2	6.87	7.30	5.89
	3	6.75	7.28	7.23
	4	6.78	7.28	6.86
	5	6.87	7.24	5.19
	6	7.20	7.35	1.96
B	1	7.00	7.31	4.23
	2	6.96	7.21	3.53
	3	6.85	7.12	3.70
	4	6.94	7.16	3.14
	5	7.08	7.20	1.61
	6	7.24	7.25	0.11

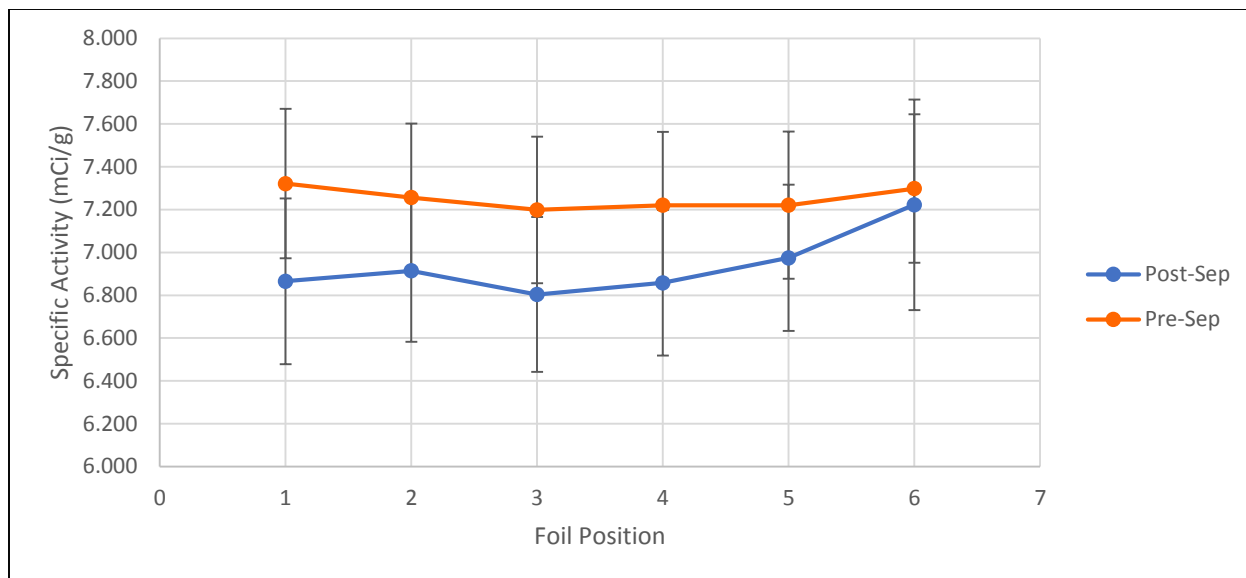


Figure 21: Average Specific Activities between Foil Sets

4.3 Isochain Comparisons

Table 6 and 7 display the significant percent difference between the activity calculated by IsoChain and the data collected from the samples prior to chemical separation and after chemical separation, respectively. The decay per second (DPS) for these samples is displayed rather than the specific activity because the specific activity for every sample would be the same for the IsoChain calculations, as the code does not compensate for flux depression.

The results for the post separation were consistently higher than those for the pre-separation, which was to be expected due to loss of material in the columns. Another feature that stood out about these results is the approximately 30% difference in the theoretical IsoChain calculations and the experimental values. Research into the cause of this phenomena needs to be undertaken in the future.

Table 6: Pre-Separation Experimental vs. Isochain Results

Target Set	Target Position	Experimental DPS { $\times 10^7$ }	IsoChain DPS { $\times 10^7$ }	% Difference
A	1	2.22	3.20	30.6
	2	1.75	2.60	32.7
	3	1.92	2.79	31.1
	4	1.94	2.82	31.1
	5	2.04	2.97	31.5
	6	2.14	3.08	30.5
B	1	1.92	2.77	30.8
	2	2.17	3.17	31.7
	3	1.98	2.93	32.6
	4	2.09	3.08	32.2
	5	1.86	2.77	32.7
	6	1.84	2.61	29.8

Table 7: Post-Separation Experimental vs. Isochain Results

Target Set	Target Position	Experimental DPS { $\times 10^7$ }	IsoChain DPS { $\times 10^7$ }	% Difference
A	1	2.04	3.20	36.2
	2	1.69	2.60	35.0
	3	1.78	2.79	34.0
	4	1.81	2.82	35.9
	5	1.93	2.97	35.0
	6	2.10	3.08	31.4
B	1	1.84	2.77	33.7
	2	2.09	3.17	34.1
	3	1.90	2.93	35.1
	4	2.03	3.08	34.3
	5	1.85	2.77	33.0
	6	1.72	2.61	31.6

4.4 MCNP Comparison

MCNP was used to either verify or discredit the trends witnessed in the experimental results. Figures 21 and 22 show the track length estimate of a cell flux (f4 Tally) across each of the foils for thermal neutrons and neutrons of all energies, respectively. The plots are arbitrarily labeled as the “upper” and “lower” targets because the distinction of which set of foils (A or B) was on top and which was on bottom within the aluminum rabbit placed in HFIR was not specified in Spencer Walsh’s notes.

It is important to know that the actual values of these results are arbitrary, as they vary with the number of allowed particle simulations in MCNP. The program was simply allowed to run until it produced results with a relative error less than 1% because calculating the exact number of neutrons that were produced in the reactor for the two hours irradiation of these targets were in it is beyond the scope of this project. The important feature of these plots is relationship between the foils, as the flux of neutrons to the target should be isotropic, and any variations between the foils could be accounted to neutron depression. However, as the plots show, there is very little variation.

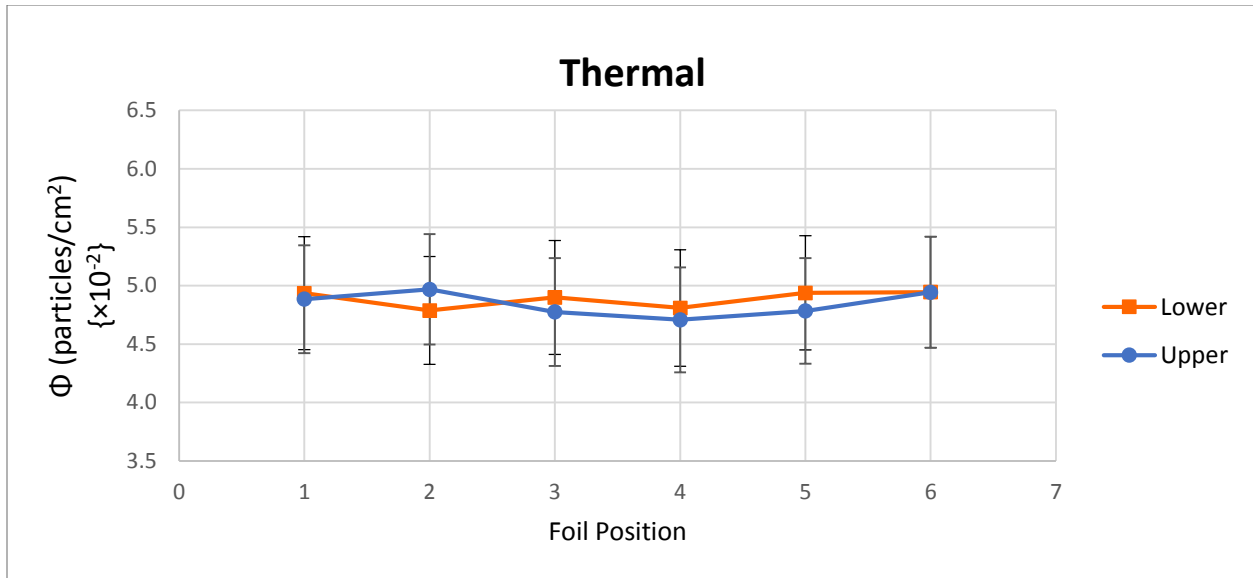


Figure 22: MCNP Cell Flux Values for Lower Target

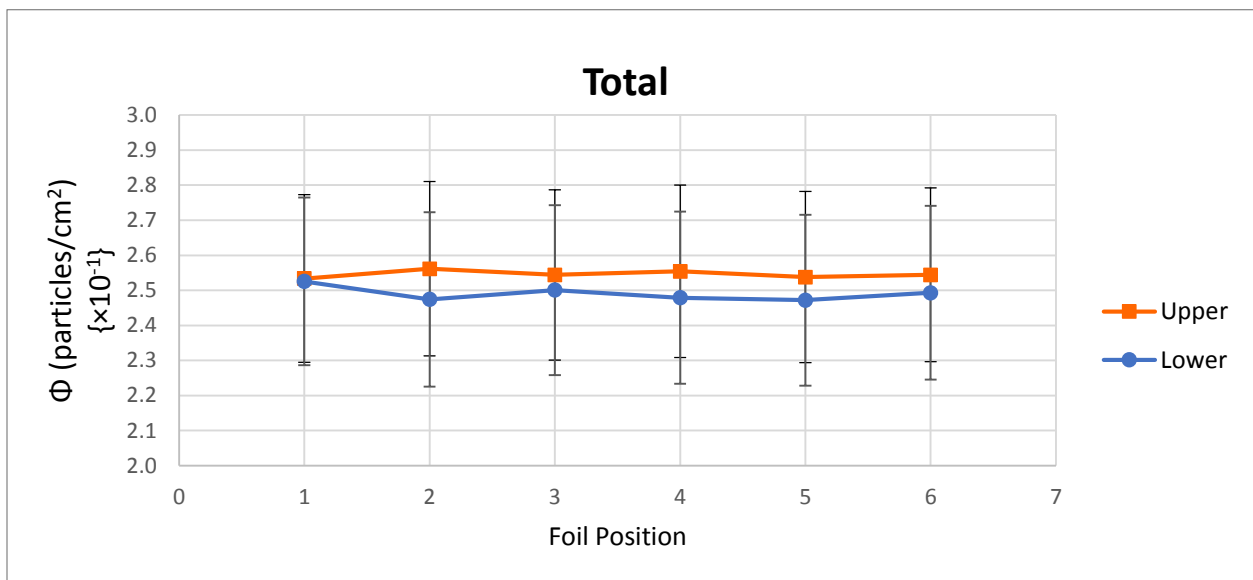


Figure 23: MCNP Cell Flux Values for Lower Target

Chapter 5: Conclusions

Although there is merit to the underlying theory of this method for producing nuclides with higher specific activity, the data from this experiment cannot verify its practical effectiveness. Due to the very small loss of specific activity witnessed between some samples' pre-separation and post-separation results, the contribution of the ^{60}Co and ^{154}Eu to the liquid scintillation count rate can be considered marginal for these samples. Because of this, the fact that the pre-separation results are so linear contradicts the theory of neutron flux depression. It is questionable if even the approximately 5% increase seen in the post-separation results merit justification for altering the standard geometry of targets within HFIR, especially if that variation could possibly be attributed to losses throughout the ion exchange chromatography procedure. However, this data does, albeit marginally, support the theory of altering target geometry to increase specific activity.

Of course, there are a variety of possible causes as to why the expected trend for neutron depression was only slightly noticeable in these samples. The most likely cause is the relatively small absorption cross sections in ^{62}Ni , which simply makes neutron activation in the foils unlikely. Other possible sources for the similarity of specific activities between the foils include poor target positioning/experiment design, as the neutron flux was able to directly strike every foil from the sides not touched by the other foils. Because of the incredibly small target size and relatively short irradiation time, very little ^{63}Ni was produced in each sample; therefore, this experiment in its essence was extremely delicate, requiring the utmost care and effort in ensuring the avoidance of cross contamination and loss of material. Although great lengths were taken to avoid these pitfalls, as mentioned in the chemical processing procedure, a great deal of statistical uncertainty was evident in the results. However, it was found that the main contributing factor to the uncertainties seen in the results were due to the error associated with the standard supplied by Eckert and Zeigler. Improvements need to be made to the verification of the standard's activity in order to truly validate this research.

If future work is to be done regarding this project, hopefully the procedure for the measurement and chemical processing of ^{63}Ni targets described in this thesis will be of

significant help to the next researcher. More immediately, however, work could be done to calculate a theoretical absorption cross-section using MCNP's residual nuclei tally (f8) and compare it to the evaluated data presented in Mughabgab as well an experimentally acquired value from this data. Additionally, it would be beneficial to investigate cause for the discrepancies between IsoChain values and the experimental values. Of course, if this project were to be reproduced in the future with the objective of investigating neutron depression in the foils, it would be ideal to alter the design of the experiment, placing the target foils in a uniform energy neutron beam rather than the flux trap of HFIR, in order to minimize the uncertainty of the results. However, regarding the production of isotopes via neutron bombardment within a reactor, this research would not be entirely applicable.

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Appendices

Appendix A: IsoChain Calculations

Table A.1: IsoChain Calculated Sample Activity

	EOB Activity (Bq) { $\times 10^7$ }	Current Activity (Bq) { $\times 10^7$ }
1g	40.56	38.77
A1	3.35	3.20
A2	2.72	2.60
A3	2.92	2.79
A4	2.95	2.82
A5	3.11	2.97
A6	3.22	3.08
B1	2.90	2.77
B2	3.32	3.17
B3	3.07	2.93
B4	3.22	3.08
B5	2.89	2.76
B6	2.73	2.61

Note: The “Current Activity” is the activity calculated compensating for exponential decay using a half-life of 101.2 years to the date 2/13/19.

Appendix B: Foil LSC Measurements

Table B.1: Pre-Separation LSC Measurements

Target Set	Target Position	Cts-Bkg { $\times 10^5$ }	Activity (Ci) { $\times 10^{-4}$ }	Measured Mass (mg)	Specific Activity (mCi/g Ni)	Total Uncertainty (%)
A	1	1.5	6.0	81.95	7.34	3.4
	2	1.2	4.9	66.58	7.30	3.4
	3	1.3	5.2	71.37	7.28	3.4
	4	1.3	5.3	72.17	7.28	3.4
	5	1.4	5.5	76.06	7.24	3.4
	6	1.5	5.8	78.86	7.35	3.4
B	1	1.3	5.2	70.97	7.31	3.4
	2	1.5	5.9	81.16	7.21	3.4
	3	1.3	5.3	75.07	7.12	3.4
	4	1.4	5.7	78.86	7.16	3.4
	5	1.3	5.1	70.77	7.20	3.4
	6	1.2	4.8	66.88	7.25	3.4

Table B.2: Post-Separation LSC Measurements and Calculations: A1-A6

Target Position	Cts-Bkg { $\times 10^5$ }	Activity (Ci) { $\times 10^{-4}$ }	Measured Mass (mg)	Specific Activity (mCi/g Ni)	Total Uncertainty (%)
A1.1	1.3	5.60	81.95	6.83	3.4
A1.2	1.3	5.52	81.95	6.73	3.4
A1.3	1.3	5.44	81.95	6.64	3.4
A2.1	1.1	4.58	66.58	6.87	3.4
A2.2	1.1	4.58	66.58	6.87	3.4
A2.3	1.1	4.56	66.58	6.86	3.4
A3.1	1.2	4.84	71.37	6.78	3.4
A3.2	1.2	4.88	71.37	6.84	3.4
A3.3	1.1	4.74	71.37	6.64	3.4
A4.1	1.2	4.86	72.17	6.73	3.4
A4.2	1.2	4.88	72.17	6.77	3.4
A4.3	1.2	4.93	72.17	6.83	3.4
A5.1	1.3	5.25	76.06	6.91	3.4
A5.2	1.3	5.22	76.06	6.86	3.4
A5.3	1.3	5.20	76.06	6.83	3.4
A6.1	1.3	5.38	78.86	6.82	3.4
A6.2	1.4	5.78	78.86	7.33	3.4
A6.3	1.5	5.95	78.86	7.55	3.4

Table B.3: Post-Separation LSC Measurements and Calculations: B1-B6

Target Position	Cts-Bkg { $\times 10^5$ }	Activity (Ci) { $\times 10^{-4}$ }	Measured Mass (mg)	Specific Activity (mCi/g Ni)	Total Uncertainty (%)
B1.1	1.2	4.96	70.97	6.99	3.4
B1.2	1.2	4.95	70.97	6.98	3.4
B1.3	1.2	4.99	70.97	7.04	3.4
B2.1	1.4	5.65	81.16	6.96	3.4
B2.2	1.4	5.69	81.16	7.01	3.4
B2.3	1.4	5.62	81.16	6.92	3.4
B3.1	1.3	5.13	75.07	6.83	3.5
B3.2	1.3	5.15	75.07	6.86	3.4
B3.3	1.3	5.16	75.07	6.88	3.4
B4.1	1.3	5.38	78.86	6.82	3.4
B4.2	1.4	5.54	78.86	7.02	3.4
B4.3	1.4	5.50	78.86	6.98	3.4
B5.1	1.2	5.06	70.77	7.14	3.4
B5.2	1.2	5.05	70.77	7.13	3.4
B5.3	1.2	4.94	70.77	6.97	3.4
B6.1	1.2	4.69	66.88	7.01	3.4
B6.2	1.2	4.72	66.88	7.05	3.4
B6.3	1.2	4.71	66.88	7.04	3.4

The pre-separation and post-separation raw data can be found as ATTACHMENT 6 and ATTACHMENT 7, respectively.

Appendix C: MCNP Input

Because the length of code required for this run, the code for the MCNP calculations can be found in the List of Attachments section of this report. [ATTACHMENT 5]

Vita

Zane Wallen is the second son of Julie and Christopher Wallen and was born and raised in eastern Tennessee. He graduated from Jefferson County High School in 2012 and received his Bachelor's Degree in Nuclear Engineering with a concentration in Radiological Engineering from the University of Tennessee in the spring of 2017. He is currently attending University of Tennessee to achieve a Master in Science in Nuclear Engineering with a concentration in Radiological Engineering as well.