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Nuclear Data of Proton-Induced Reactions for Accelerator-Based Isotope Production

by

Yun-Hsuan “Abby” Lee

A dissertation submitted in partial satisfaction of the

requirements for the degree of

Doctor of Philosophy

in

Engineering – Nuclear Engineering

in the

Graduate Division

of the

University of California, Berkeley

Committee in charge:

Professor Lee Bernstein, Chair

Professor Barbara Jacak

Professor Daniel Siefman

Professor Karl Van Bibber

Spring 2026

Nuclear Data of Proton-Induced Reactions for Accelerator-Based Isotope Production

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Yun-Hsuan “Abby” Lee

Abstract

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Doctor of Philosophy in Engineering – Nuclear Engineering

University of California, Berkeley

Professor Lee Bernstein, Chair

This dissertation includes two works of the Tri-laboratory Effort in Nuclear Data (TREND), a collaboration among Lawrence Berkeley National Laboratory, Los Alamos National Laboratory, and Brookhaven National Laboratory that undertakes the challenges in accelerator-based isotope production. The experiments include the continuation of a long-standing campaign of cross section measurements via stacked-target irradiations, and a venture into the realm of in-beam γ -ray spectroscopy, rarely employed in the field of isotope production.

Motivated by the demand of ^{103}Pd for brachytherapy, characterization of the $^{\text{nat}}\text{Ag}(\text{p},\text{x})$ reactions between incident proton energy of 40 MeV and 200 MeV was performed. The experimental methods and results of stacked-target irradiations on targets of silver, copper, and nickel are discussed, with an additional focus on the underlying complications in secondary-particle-induced reactions and the usage of monitor reactions. Experimental measurements made were used to further the data evaluation methodologies developed by the TREND collaboration via the investigation of parameterized charged-particle reaction modeling with TALYS-2.0.

To address the need for $^{202\text{g}}\text{Pb}$ of the geochronology community, the TREND collaboration pivoted to the experimental techniques of in-beam γ -ray spectroscopy at the 88-Inch Cyclotron of Lawrence Berkeley National Laboratory. With a long half-life and no observable γ -ray emission following electron capture decay, the quantification of $^{202\text{g}}\text{Pb}$ production was not possible through activation analysis. This work provides the first ever direct measurement of the $^{\text{nat}}\text{Tl}(\text{p},\text{x})^{202\text{g}}\text{Pb}$ reaction at proton energies 30 and 50 MeV. New insights into the structure of ^{202}Pb and additional measurements of γ -ray production cross sections in lead isotopes with $198 \leq A \leq 203$ provided an unique opportunity in the exploration of effects of angular momentum in level density modeling, where conclusions suggested that the overestimation of the widths of angular momentum distributions could be masked by manipulations to the optical model potential.

Efforts in the isotope production field have long been hindered by uncertainties in modeling-based predictions, particularly for reactions with high-energy charged incident particles. In generating previously unmeasured experimental charged-particle reaction cross section data and improving the understanding in the phenomenology of charged-particle reaction modeling, this dissertation is an essential enhancement in both completeness and quality of nuclear data for accelerator-based isotope production.

可知世上萬般，好便是了，了便是好。

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Chapter 1

Introduction

1.1 Nuclear Data in Isotope Production

Before nuclear technologies can be achieved, nuclear data for the relevant isotopes must be obtained. This includes information such as masses, half-lives, structures, decay radiations, and well-quantified descriptions of nuclear reactions of interest. The developments in isotope production are often constrained by the significant knowledge gaps in reaction cross section data, particularly of charged-particle-induced reactions.

The identification of isotopes suitable for specific applications builds on information of decay properties – finding suitable combinations of half-lives and outgoing particle types, energies, and intensities. For instance, the use of isotopes in targeted therapy calls for nuclides with short-range decay particle emissions and few high-energy decay γ -rays. However, after an isotope of interest is identified and prioritized, crucial challenges arise from designing optimized production pathways. The lack of production cross section measurements often hinders the search of reactions that produce isotopes of interest in sufficiently high quantities and purities.

1.1.1 Accelerator-Based Isotope Production

Isotopes, both radioactive and stable, are routinely used for varying applications from cancer diagnosis and treatment to environmental assays, geochronology, and space exploration. The isotopes, each with unique nuclear properties fitting to its purpose, are regularly produced at particle accelerators or nuclear reactors around the world.

With the advancements in medical applications of isotope production, the use of artificial radioactivity is steadily increasing worldwide. One of the most common medical isotopes today remains to be $^{99\text{m}}\text{Tc}$, for its use in SPECT (single-photon emission computed tomography). $^{99\text{m}}\text{Tc}$ is extracted from the decay of its parent nuclide, ^{99}Mo , which itself is mostly produced at nuclear reactors. Despite its early realization and commonality, reactor-based isotope production is limited to neutron-induced reactions, creating challenges for the production of high specific activity samples needed for medical use. This largely restricts the

possibility in reaction routes and hence product isotopes. Other associated concerns include fuel material proliferation and general reactor safety.

Every day, charged-particle accelerators produce a vast selection of isotopes for PET (positron emission tomography) scans, targeted radionuclide therapy, *etc.* Ongoing research in the use of Auger-Meitner electron (*e.g.*, ^{103}Pd) or α -particle (*e.g.*, ^{225}Ac) emitters promise even higher effectiveness and precision in tumor-cell killing than the currently-in-use β -particle emitters (*e.g.*, ^{177}Lu). Accelerator-based isotope production allows for not only higher flexibility in reaction pathways, but also the possibility of on-site production of short-lived isotopes. ^{18}F ($t_{1/2} = 109.734(8)$ min) is an example of those produced at low-energy cyclotrons scattered globally at medical centers.

Isotope production also supports improvements and discoveries in fields such as geology, nuclear battery design, environmental studies, national security, and radiation detection. Missions of the isotope production field include addressing shortages in the domestic stable/long-lived isotope inventories. ^{202}gPb , whose production is assessed in this dissertation, is one of the isotopes proposed to be produced at accelerators due to its demand in the field of geochronology using precision mass spectroscopy.

Domestic regional high-energy and intensity charged-particle accelerators, such as IPF (Isotope Production Facility) at LANL (Los Alamos National Laboratory) and BLIP (Brookhaven Linac Isotope Producer) at BNL (Brookhaven National Laboratory), open up potential for larger-scale production and access to production pathways with higher reaction thresholds. In addition to charged particles, neutrons are also available at accelerators via deuteron breakup and spallation.

While isotope production is facilitating numerous physics applications and advancing approaches to combating illnesses, further breakthrough is obstructed by our lack of understanding in fundamental nuclear phenomena.

1.1.2 Nuclear Data For Proton-Induced Reactions

The origin of the nuclear physics field naturally led to an abundance of data of neutron-induced reactions. With weapons and reactors pushing the science forward, nuclear data measurements have been dominated by neutron experiments; even though the number of charged-particle-induced reaction measurements has increased significantly since the 1970s, the imbalance in experimental nuclear data availability between neutron- and charged-particle-induced reactions still exists to this day [1].

Beyond the compilation of experimental reaction data, such as the Experimental Nuclear Reaction Data Library (EXFOR) [1], it is the evaluation of nuclear data that ultimately creates suggested values that should be adopted in nuclear application designs. Nuclear data evaluation encompasses careful assessments of all compiled measured data, from experimental configurations to uncertainty assignments, and intensive and iterative modeling using established theories. Such evaluations are found in libraries such as the Evaluated Nuclear Data File (ENDF) [2], the US-based evaluated nuclear reaction library, where the discrepancy between numbers of entries of neutron and charged-particle data further increases. In

ENDF/B-VIII.0, released in 2018, there exist 527 neutron evaluations yet only 49 proton evaluations [2].

The scarcity of non-evaluated and evaluated data for proton-induced reactions is even more dire at relatively higher proton energies, particularly above ≈ 100 MeV. This consequently raises the difficulty in developing accurate low-energy nuclear reaction models and hence effective predictive tools of reaction cross sections. The determination of optimal production routes and engineering designs in isotope production relies on the accuracy of yield predictions of *all* possible reaction products, including both the isotopes-of-interest and the co-produced. Due to the limitations of chemical separation, co-production of isotopes of the same element to the isotope-of-interest is of particular concern. As a result, there must be a generally applicable reaction model that covers the wide variability in nuclide and reaction properties for isotope production planning. Although phenomenological reaction modeling programs such as Talys have long been established, there still exist significant uncertainties in the accuracies of these reaction cross section calculations. The increase in experimental measurements is essential to refine the parameterization of such tools.

Due to the current lack of confidence for reaction simulations, reaction cross sections must be measured every time a new production pathway is explored. To meet the nuclear data needs for the emerging novelties and ever-evolving techniques in isotope production, not only should more experiments be conducted, but the capabilities of reaction modeling and nuclear data evaluation must also be enhanced.

1.2 Goal of This Work

This dissertation includes two nuclear data measurements for prioritized isotope production pathways, as part of the TREND (Tri-Laboratory Effort in Nuclear Data) and TREND 2.0 efforts. The experiments, performed at three charged-particle accelerators, generate extensive proton-induced reaction cross section data up to 200 MeV. Specifically, this work spans measurements of proton-induced reactions on silver, copper, nickel, and thallium.

This work further explores reaction theories and modeling in the phenomenological reaction modeling package Talys informed by the experimental cross section data. This aims to improve general prediction capabilities in charged-particle reaction modeling and facilitate future efforts in isotope production.

The main body of this dissertation is comprised of two chapters, each exploring the measurement and modeling of proton-induced reactions on a natural target. The two experiments are performed via distinct methods and motivated by different applications.

Chapter 2 of this dissertation details the investigation of the $^{\text{nat}}\text{Ag}(p,x)$ reaction route for the production of ^{103}Pd . Performed via the stacked-target method, this is the first measurement to be conducted for the $^{\text{nat}}\text{Ag}(p,x)^{103}\text{Pd}$ reaction above 100 MeV. In addition, energy-differential cross section data for 24 $^{\text{nat}}\text{Ag}(p,x)$ reactions, 23 $^{\text{nat}}\text{Cu}(p,x)$ reactions, and 16 $^{\text{nat}}\text{Ni}(p,x)$ reactions are reported. Cross section data free of secondary-particle-induced production contamination are further used for the exploration of Talys parameters. This

follows the continuing efforts of the TREND collaboration in investigations of nuclear level densities, optical potentials, and pre-equilibrium reaction modeling [3, 4, 5, 6, 7].

Chapter 3 discusses the production of $^{202\text{g}}\text{Pb}$ via proton bombardment on natural thallium. Due to the long half-life and the lack of γ -rays following the decay of $^{202\text{g}}\text{Pb}$, this experiment ventures beyond the common practices in isotope production of solely relying on stacked-target irradiations and decay gamma spectroscopy and pivots to the use of prompt gamma spectroscopy. Direct measurement of angular- and energy-differential cross section ratios is reported for product lead nuclei of mass numbers between 198 and 203. The prompt γ -ray analysis includes recommended changes to the evaluated nuclear structure data of $^{202\text{g}}\text{Pb}$. As the newest evolution of the study of Talys modeling inputs, insight into the population of specific states provides the exploration of reaction modeling with improved accuracy in spin-parity-dependent parameters.

This dissertation forwards the field of isotope production not only in the generation of previously unmeasured nuclear data, but also the expansion of fundamental experimental methods via new adaptations and the understanding of underlying reaction theories.

Chapter 2

Measurement and Modeling of Proton-Induced Reactions on Silver from 40 to 200 MeV

2.1 Abstract

The lack of cross section measurements has been hindering the isotope production efforts at accelerator facilities with 100 MeV or higher energy proton beams. The TREND (Tri-Laboratory Effort in Nuclear Data) 2.0 collaboration was formed between Lawrence Berkeley National Laboratory (LBNL), Los Alamos National Laboratory (LANL), Brookhaven National Laboratory (BNL), and Texas A&M University to perform measurements of high-priority isotope production reaction routes and further the prediction capabilities of reaction modeling. For this work, stacked-targets consisting of natural copper, nickel, and silver foils were irradiated at the BNL Brookhaven LINAC Isotope Producer (BLIP) and the LANL Isotope Production Facility (IPF) to measure the cross sections of $^{\text{nat}}\text{Ag}(\text{p},\text{x})$ reactions between 40 and 200 MeV. This dataset includes first measurements of multiple $^{\text{nat}}\text{Ag}(\text{p},\text{x})$ reactions at above 100 MeV, including that of $^{\text{nat}}\text{Ag}(\text{p},\text{x})^{103}\text{Pd}$. Production cross sections are reported for an additional 24 $^{\text{nat}}\text{Ag}(\text{p},\text{x})$ reactions, 23 $^{\text{nat}}\text{Cu}(\text{p},\text{x})$ reactions, and 16 $^{\text{nat}}\text{Ni}(\text{p},\text{x})$ reactions. Measured cross sections are compared with literature data and theoretical calculations from Talys-2.0. Building on past efforts in enhancing predictive capabilities of charged-particle-induced reaction cross sections, a parameter optimization process was applied for the level density, photon strength, optical model potential, and two-component exciton model for pre-equilibrium reactions in Talys-2.0. This resulted in overall improved modeling of $^{\text{nat}}\text{Ag}(\text{p},\text{x})$ reaction cross sections compared to default calculations.

2.2 Introduction

^{103}Pd ($t_{1/2} = 16.991(19)$ d) decays solely by electron capture, followed by the emission of X-rays and Auger-Meitner electrons. The most intense γ -ray in its decay is of $E_\gamma = 39.7$ keV and $I_\gamma = 0.0683\%$ [8]. Such decay properties are favored by the medical community for applications in brachytherapy, during which pellets or plaques containing radioisotopes are directly placed near tumor sites. Brachytherapy is successful in treating localized cancers on or close to the surface of the human body, as ^{103}Pd treatments have successfully demonstrated. For instance, the effectiveness of ^{103}Pd brachytherapy in the treatment of choroidal melanoma and prostate cancer has been published [9, 10]. The potential of ^{103}Pd in treatment has long been realized, yet the search for its optimal production route has not been concluded.

Explored reactions for ^{103}Pd production, in addition to $^{\text{nat}}\text{Ag}(p,x)$ discussed in this work, include $^{102}\text{Pd}(n,\gamma)$, $^{102}\text{Rh}(p,n)$, and $^{102}\text{Rh}(d,2n)$. However, concerns about efficiency in production, purity of product, specific activities, and ultimately the high cost of enriched targets motivate further research into more feasible production routes [11].

There currently only exists two literature cross section datasets of $^{\text{nat}}\text{Ag}(p,x)^{103}\text{Pd}$ which cover proton energies up to 100 MeV [11, 12]. Not only does the complete lack of information in the reaction above 100 MeV impede possible production at facilities such as BNL-BLIP ($E_p = 200$ MeV), the disagreement between the datasets further complicates the process of production planning.

Where empirical cross section data are not available, limited or widely discrepant, data evaluations are not possible, and theoretical calculations are notoriously challenging. To fill in the gaps in the experimental data, important efforts are being made to establish simulated nuclear data libraries to address the needs of all nuclear science application R&Ds. One of the major libraries is TENDL, built on the capabilities of Talys, a phenomenological nuclear reaction modeling program with tunable parameters [13]. Nonetheless, it has been shown in past publications that Talys typically fails to predict cross section values, especially at above 100 MeV [3, 4, 5, 6, 7]. The lack of a comprehensive theory of low-energy nuclear physics leads to a reliance on phenomenological simulations that can only be truly refined through the addition of new experimental data.

2.3 Experimental Methods and Materials

The facilities at which this work was performed include BNL-BLIP for $100 < E_p < 200$ MeV and LANL-IPF for $40 < E_p < 100$ MeV. One set of foils is irradiated at each accelerator; they will hereafter be referred to as the “BNL stack” and the “LANL stack”.

2.3.1 Stacked-Target Design

The stacked-target (also known as stacked-foil) method was adopted for this work. By activating a stack consisting of thin foils and thick degraders with a well-characterized charged particle beam, multiple energy-differential cross sections can be simultaneously measured for each observed reaction pathway. Monitor foils were added to the stack to ensure accurate measurement of beam energy and intensity degradation of the beam through the stack. The stacked-target schematic is shown in Figure 2.1.

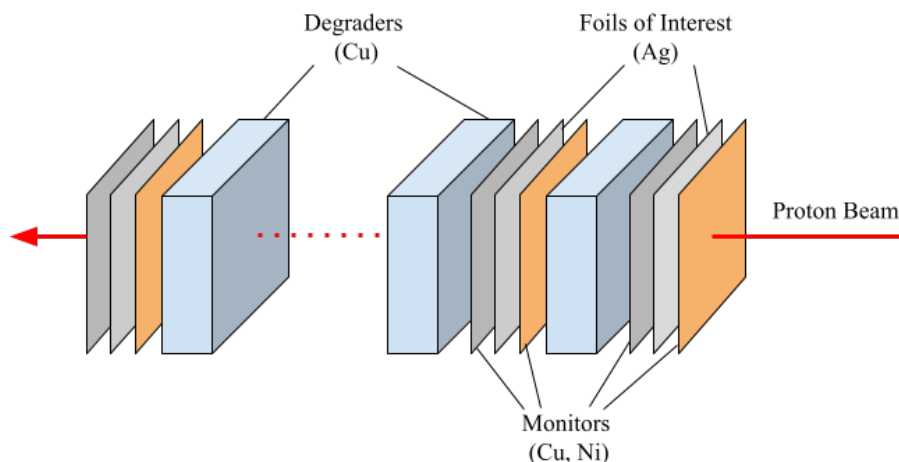


Figure 2.1: The stacked-target configuration.

BNL Stack and Irradiation

The BNL stack consisted of 7 natural silver, copper, and nickel foils each. All foils were of $25\text{ }\mu\text{m}$ nominal thickness, purchased from Goodfellow Corporation (Coraopolis, PA 15108, USA). Each foil was purchased as or cut into a 2.5 cm by 2.5 cm square then cleaned with isopropyl alcohol. The length and width of each foil were measured with a digital caliper (Mitutoyo America Corp.), and the thickness was measured with a digital micrometer (Mitutoyo America Corp.) to $1\text{-}\mu\text{m}$ precision. Each foil was then weighed to 0.0001-g precision. The areal densities of the foils were all of $\leq 0.1\%$ uncertainty.

The foils were suspended with 3M Polyimide Film Tape 5413 (Kapton) in acrylic frames. The beam-strike areas on the foils were not covered with tape. Each silver foil was paired with a copper foil and a nickel foil; the three foils were then secured together into a set with a bailing wire, as shown in Figure 2.2. A copper degrader was added between each set of foils. The dimensions of the degraders were measured as for the foils, and they were weighed to 0.01-g precision. Finally, a stainless foil of nominal thickness 0.1 mm each was added to the front and back of the stack. The stainless steel foils served to confirm, post-irradiation, that the beam-strike area on each foil was well-contained within the center of the foil via

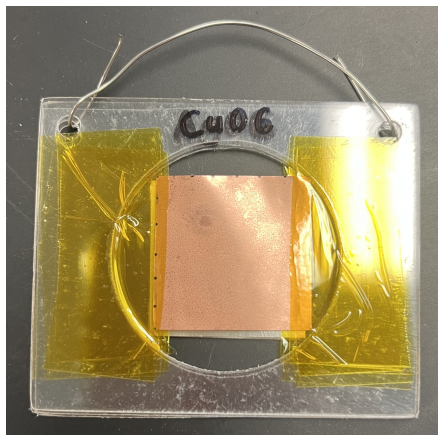


Figure 2.2: Photo of a set of copper, silver, and nickel foils connected by bailing wire.

dose-mapping onto radiochromic film (Gafchromic EBT3). The specifications of each foil and degrader in the stack can be found in Appendix A. The full assembled stack is shown in Figure 2.3.

Before the incident protons were upon the first foil of the stack, they traversed the AlBeMet (Al-Be alloy) and 316 stainless steel beamline windows and water cooling channels. The target box itself had a 316 stainless steel entrance window of 0.960 mm. All components were taken into consideration for the calculation of the incident beam current on each foil.

The BNL target stack was irradiated for 3650 s with a nominal current of 150 nA at 200 MeV on April 15, 2025.

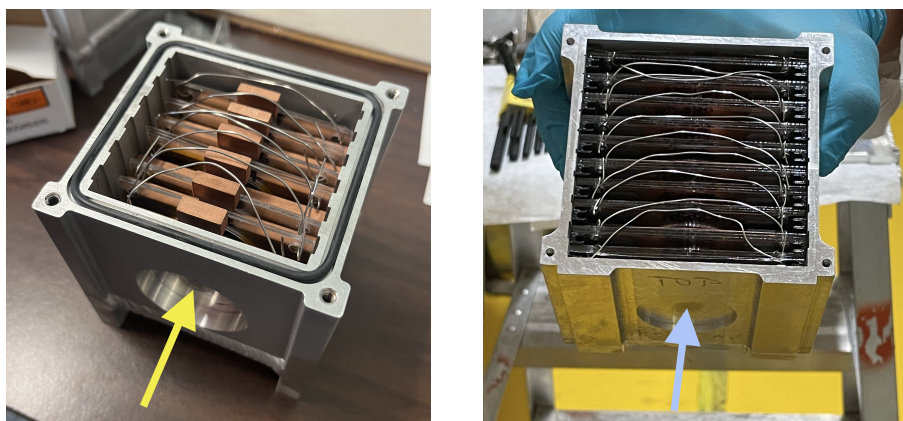


Figure 2.3: Photos of the assembled BNL stack (left) and LANL stack (right) in respective target boxes. The arrows show the direction of incident beam.

LANL Stack and Irradiation

The LANL stack included 10 natural silver, natural copper, and natural nickel foils each. All foils used in the LANL stack were purchased from Goodfellow Corporation (Coraopolis, PA 15108, USA) and had identical nominal dimensions as the foils used in the BNL stack. The same dimension and mass measurements were performed as for the BNL stack. Similarly, the areal densities of all foils were of $\leq 0.1\%$ uncertainty. As in the case for the BNL stack, each silver foil was paired up and connected by bailing wire to a copper foil and a nickel foil after all foils were suspended to acrylic frames with Polymide Film Tape 5413 (Kapton). In between each set of foils, a thick copper degrader was inserted. A stainless steel foil of nominal thickness $50\text{ }\mu\text{m}$ was added each at the front and back of the stack from beam profile monitoring. Following irradiation, radiochromic films were placed on the stainless steel foils where they were exposed to decay radiation (primarily β -particles) and used to provide a spatial profile of the beam. The activated radiochromic films are shown in Figure 2.4. The full assembled stacked is shown in Figure 2.3.

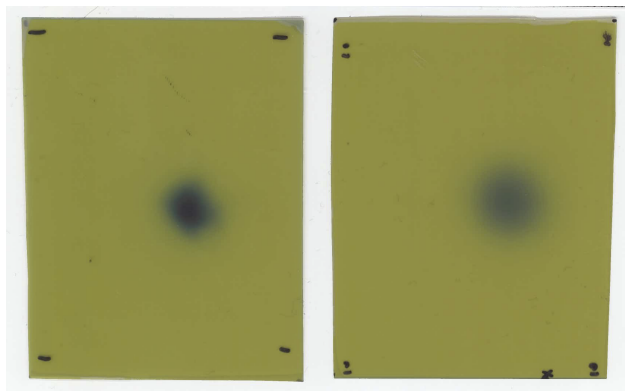


Figure 2.4: The dark areas on the films represent the beam-strike areas on the stainless steel foils. As seen, the beam-strike area remained well-contained within the dimensions of the foils (identical to that of the films) from the front (left) to the back (right) of the stack.

Before the proton beam was seen by the first foil of the stack, the beam traverses an Inconel 718 beam window and a water cooling channel. The target box itself had an aluminum entrance window of thickness 0.254 mm . All components were taken into consideration for the calculation of the incident beam current on each foil.

The LANL stack was irradiated for 3601 s with a nominal current of 200 nA at 100 MeV on August 19, 2025.

2.3.2 Decay Gamma Spectroscopy and Activity Measurements

BNL Data Acquisition

The counting of γ -rays started at BNL-BLIP approximately 1 hour after the end-of-bombardment (EoB). Detectors used at BNL include 2 ORTEC tran-Spec detectors, an ORTEC IDM-200-V HPGe detector, and an ORTEC GLP-36360/13-P-S low-energy radiation detector. The detectors at BNL were calibrated with ^{109}Cd , ^{133}Ba , ^{137}Cs , ^{152}Eu , and ^{241}Am sources (Eckert & Ziegler, Valencia, CA 91355 USA).

Samples were shipped to LBNL after the initial 5 days of counting at BNL. Counting resumed at LBNL after 4 days of shipping and processing and were conducted for over the period of about 10 weeks. Detectors used at LBNL include an ORTEC IDM-200-V HPGe detector, an ORTEC GMX-50220-S HPGe detector, and an ORTEC GLP-36360/13-P-S low-energy radiation detector. The detectors at LBNL were calibrated with purchased ^{109}Cd , ^{133}Ba , ^{137}Cs , ^{152}Eu , and ^{241}Am sources (Eckert & Ziegler). A ^{57}Co source made in-house at LBNL was also used.

LANL Data Acquisition

Counting started at LANL-IPF approximately 2 hours after EoB. Detectors used at LANL include 2 ORTEC GEM20P4-70-PL HPGe detectors, an ORTEC GEM20P-PLUS detector, and an ORTEC GLP-36360/13-P-S low-energy radiation detector. The detectors at LANL were calibrated with ^{133}Ba , ^{137}Cs , ^{152}Eu , and ^{241}Am sources (Eckert & Ziegler).

The samples were shipped to LBNL after counting for 6 days at LANL. The counting resumed at LBNL resumed after 2 days of shipping and processing and lasted for another 4 months. At LBNL, the identical set of detectors and calibration sources were used as in the case of the BNL stack.

Activation Analysis

The CURIE python library [14] was used for the analysis of all collected spectra, including peak-fitting and detection calibrations in energy, efficiency, and resolution. CURIE also supports the calculation of time-averaged production rates. The procedures are well-described in Morrell *et al.* [7] and is summarized here.

For each isotope, the number of decays observed in each spectra was calculated using Equation 2.1:

$$N_D = \frac{N_C}{I_\gamma \varepsilon(E_\gamma)} \frac{t_{\text{real}}}{t_{\text{live}}} \quad (2.1)$$

where N_C is the number of net counts, t_{real} is the actual time over which the detection took place, t_{live} is the live time of detection taking into account data acquisition “dead time”, I_γ is the intensity of the γ branching ratio, and $\varepsilon(E_\gamma)$ is the detection efficiency at E_γ , the energy of the γ -ray. In the case where two isotopes share the same decay γ -ray(s) (*i.e.*, decay into the same daughter isotope) but have other measurable independent decay γ -ray(s), the

shared γ -ray(s) were excluded from the calculation of isotope activities. On the other hand, where one of the parent isotopes does not have other measurable independent decay γ -ray(s), counts contributed from the decay feeding of the other parent isotope were subtracted before calculation of activity.

The production rates were then solved for isotopes in the same decay chain, with the conversion of Bateman equations, in which the activity of an isotope is defined in terms of the activity of preceding members in the decay chain [15], with an added source term for production each into a linear system shown in Equation 2.2:

$$\bar{M} \cdot \vec{R} = \vec{N}_D \quad (2.2)$$

where $\vec{R}$ is a vector of production rates, $\vec{N}_D$ is a vector of observed number of decays, and $\bar{M}$ is a matrix of decays calculated from a decay chain of unit production rates for a single isotope in $\vec{R}$, during the time interval of detection of the associated element of $\vec{N}_D$.

2.3.3 Stack Current and Energy Determination

The beam energy and current seen by each foil in the stack were determined using monitor reactions, which have well-characterized cross section data for the energy ranges of relevance in the IAEA charged-particle cross section database for medical radioisotope production [16].

Selection of Monitor Reactions

For the BNL stack ($100 < E_p < 200$ MeV), the monitor reaction is:

- ${}^{\text{nat}}\text{Cu}(p,x){}^{58}\text{Co}$

For the LANL stack ($50 < E_p < 100$ MeV), monitor reactions include:

- ${}^{\text{nat}}\text{Cu}(p,x){}^{56}\text{Co}$
- ${}^{\text{nat}}\text{Cu}(p,x){}^{58}\text{Co}$
- ${}^{\text{nat}}\text{Cu}(p,x){}^{62}\text{Zn}$
- ${}^{\text{nat}}\text{Cu}(p,x){}^{63}\text{Zn}$
- ${}^{\text{nat}}\text{Cu}(p,x){}^{65}\text{Zn}$

While ${}^{\text{nat}}\text{Ni}(p,x)$ reactions were also intended to serve as monitor reactions, concerns regarding unquantifiable secondary-particle-induced reactions resulted in the exclusion of their use (see Section 2.4.2).

The Variance Minimization Method

With the production rate R_i of each monitor reaction product, the proton beam current I_p was calculated with Equation 2.3:

$$I_p = \frac{R_i}{(\rho r)\bar{\sigma}_i} \quad (2.3)$$

, where (ρr) is the foil areal density. The flux-averaged cross section $\bar{\sigma}_i$ was calculated using Equation 2.4:

$$\bar{\sigma}_i = \frac{\int \sigma_i(E)\phi(E)dE}{\int \phi(E)dE} \quad (2.4)$$

, where $\phi(E)$ is the proton energy spectrum determined via 1-D Monte Carlo stopping power calculations using the Anderson-Ziegler formulation implemented in CURIE [7, 14], and $\sigma_i(E)$ are IAEA-recommended cross section values [16]. This accounts for uncertainties and distributions in the proton transport through the stack. The beam currents calculated from each monitor reaction for each monitor foil were fit to linear functions of proton energies ($I = I_0 + m \cdot E$).

The “variance minimization” technique employed in this work, established by Graves *et al.* [17], had been implemented in and is well-discussed in past publications [3, 4, 5, 6, 7, 18, 19]. The technique is based on the fundamental assumption that independent calculations of I_p from each monitor reaction should agree for the same foil. As shown in Figure 2.5, disagreements in I_p values could result in more than 15% relative uncertainties in beam current assignments. The technique applies a global multiplier to the areal density of all stack components, which is then varied to minimize a chosen goodness-of-fit parameter; the reduced- χ^2 is chosen for this work. The intention is not to suggest that the disagreement in I_p values stem from unknowns in foil characterization, but to account for systematic errors in proton transport calculations. The optimizing multiplier for density was -3.2% for the BNL stack and +8.70% for the LANL stack. Figure 2.5 shows the significance of the technique especially in addressing disagreements in I_p in the rear end of the stack, where the uncertainties in stopping power, proton energy straggling, and the characterization of all upstream components were amplified.

2.3.4 Cross Section Determination

Given the production rate R_i , areal density (ρr) of foil, and beam current I_p , the cross sections were calculated with Equation 2.5:

$$\sigma_i = \frac{R_i}{(\rho r)I_p} \quad (2.5)$$

Given that the derivation of beam current I_p was performed through monitor foil activation and the subsequent process of “variance minimization”, there is covariance across all cross section measurements presented in this work. The covariance is, however, not accounted for here, but should be explored in future efforts.

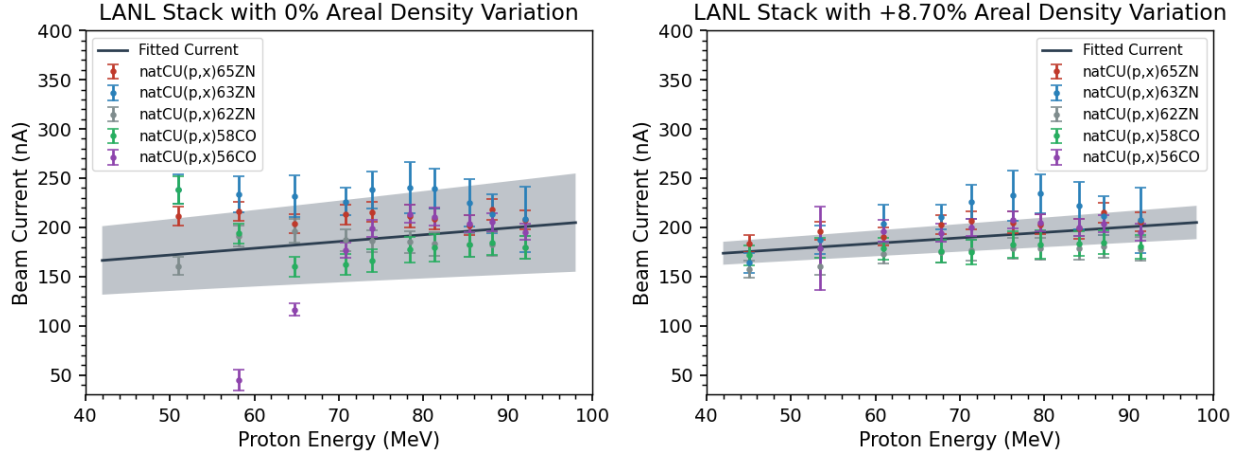


Figure 2.5: Beam current fitting before (left) and after (right) variance minimization.

The $^{\text{nat}}\text{Ag}(p,x)$ cross section results are given in Table C.1 of Appendix C, which includes that of ^{107}Cd , $^{106\text{m}}\text{Ag}$, ^{105}Ag , ^{104}Cd , ^{104}Ag , ^{103}Ag , ^{103}Pd , ^{103}Ru , ^{102}Rh , ^{101}Pd , $^{101\text{m}}\text{Rh}$, ^{100}Pd , ^{100}Rh , $^{99\text{m}}\text{Rh}$, ^{97}Rh , ^{97}Ru , ^{96}Tc , ^{95}Ru , ^{95}Tc , ^{94}Ru , $^{94\text{m}}\text{Tc}$, ^{93}Mo , ^{90}Mo , and ^{90}Nb . Tabulated cross section data for $^{\text{nat}}\text{Cu}(p,x)$ and $^{\text{nat}}\text{Ni}(p,x)$ reactions are also included in Appendix C. All decay data used in the analysis of both silver foil and monitor foil products are listed in Appendix B.

The distinction between cumulative (c) and independent (i) cross sections is made in this work. Independent cross sections are reported for isotopes that can only be directly produced via the incident proton beam or when decay-feeding from parent isotopes can be taken into account. Cumulative cross sections, which include contributions from multiple nuclear reactions, are otherwise reported. In all instances in this work, subscripts (c) and (i) indicate cumulative and independent respectively. In cases where the cross section is independent from feeding other products but not from a short-lived isomer, the product is noted with subscript ($m+$) and referred to as “total” cross sections hereafter.

2.4 Results and Discussion

Cross section values of select reactions of particular interest to the motivation of this work or reaction modeling are discussed in this section. Plots of all other measured cross sections, including those of monitor foil reactions, are presented in Appendix C. For all decay data used in analysis, including half-lives and γ -ray energies and intensities, see Appendix B.

2.4.1 Production Cross Section Measurements

$^{\text{nat}}\text{Ag}(p,x)^{103}\text{Pd}_{(c)}$

Cumulative cross sections of $^{\text{nat}}\text{Ag}(p,x)^{103}\text{Pd}$ were measured via observation of the 39.748(8) keV decay γ -ray with intensity 0.0683% [8]. Contributions to the 39.748(8) keV transition from the decay of ^{103}Ru was subtracted during activation analysis. The uncertainties in cross section primarily stem from counting statistics due to the low energy and intensity of the γ -ray. As seen in Figure 2.6, the discrepancy between the two literature measurements by Fassbender *et al.* [11] and Uddin *et al.* [12] was not clearly resolved by this work, despite the general agreement in shape of the excitation function between the work of Fassbender *et al.* and this work. It should be noted that contrasting counting techniques were adopted among the three measurements. Fassbender *et al.* performed destructive counting with chemical separations [11]; while statistical uncertainty in γ -ray counting would be improved by the direct removal of background radiation sources, uncertainties associated with separation efficiency cannot be neglected. Uddin *et al.* solely utilized the 357.45(8) keV γ -ray with 0.0221(7)% intensity for analysis [12]; challenges in counting arose from the weakness of this γ -ray and was clearly communicated by the authors.

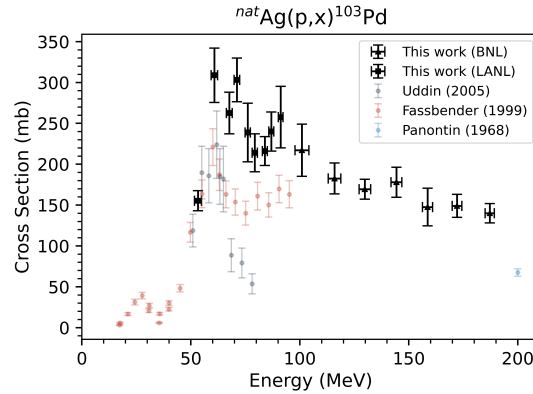


Figure 2.6: Cumulative cross section measurements of $^{\text{nat}}\text{Ag}(p,x)^{103}\text{Pd}$ [11, 12, 20]

$^{\text{nat}}\text{Ag}(p,x)^{103}\text{Ru}_{(c)}$

The cumulative cross section of $^{\text{nat}}\text{Ag}(p,x)^{103}\text{Ru}$ shown in Figure 2.7 is measured via the 497.085(10) keV γ -ray with intensity 91.0(12)%. The activities of ^{103}Ru was purposely measured despite its relatively longer half-life of 39.247(3) d due to its “contamination” of the 39.748(8) keV transition utilized in the cross section calculations of ^{103}Pd production.

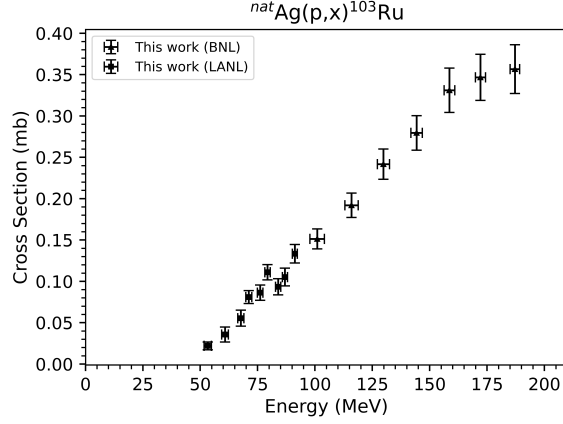


Figure 2.7: Cumulative cross section measurements of $^{\text{nat}}\text{Ag}(p,x)^{103}\text{Ru}$.

$^{\text{nat}}\text{Ag}(p,x)^{101}\text{Pd}_{(c)}$

As a co-produced Pd isotope of the $^{\text{nat}}\text{Ag}(p,x)$ production route, a good knowledge of its cumulative cross section measurements help design chemical separation protocols. As shown in Figure 2.8, results well align with both Fassbender *et al.* [11] and Uddin *et al.* [12]. The first measurements of proton energies higher than 100 MeV is reported in this work.

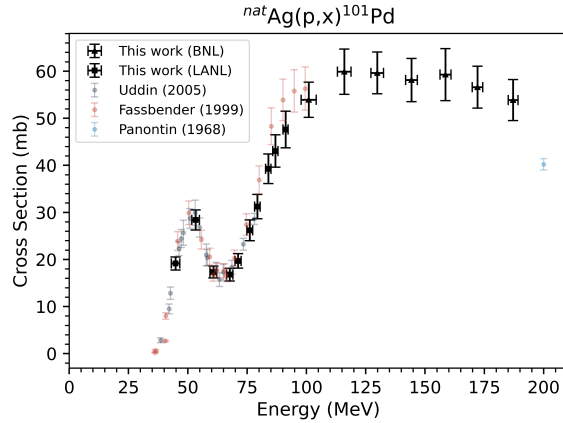


Figure 2.8: Cumulative cross section measurements of $^{\text{nat}}\text{Ag}(p,x)^{101}\text{Pd}$. [11, 12, 20]

$^{\text{nat}}\text{Ag}(p,x)^{100}\text{Pd}_{(c)}$

As another isotope of Pd, ^{100}Pd cannot be chemically separated from ^{103}Pd . With a relatively comparable half-life of $t_{1/2} = 3.63(9)$ d to ^{103}Pd , it is not feasible to simply wait for it to decay. The co-production of ^{100}Pd therefore also has to be well-quantified to inform accurate

dosage planning in brachytherapy treatments. As seen in Figure 2.9, while the shape of the excitation functions generated by Fassbender *et al.* [11], Uddin *et al.* [12], and this work generally agree, differences in magnitudes had been reported.

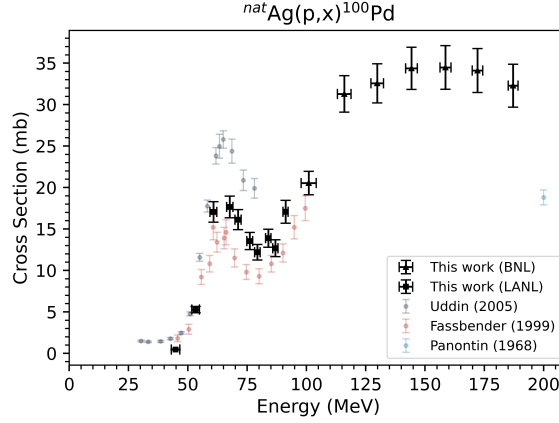


Figure 2.9: Cumulative cross section measurements of ${}^{\text{nat}}\text{Ag}(\text{p},\text{x}){}^{100}\text{Pd}$ [11, 12, 20].

${}^{\text{nat}}\text{Ag}(\text{p},\text{x}){}^{107}\text{Cd}_{(i)}$

Only produced via ${}^{107}\text{Ag}(\text{p},\text{n})$ and ${}^{109}\text{Ag}(\text{p},3\text{n})$, independent cross sections of this reaction can be reported with general agreement with literature data (Figure 2.10). Production cross sections of Cd isotopes are also all free from contamination of secondary-neutron-induced reactions.

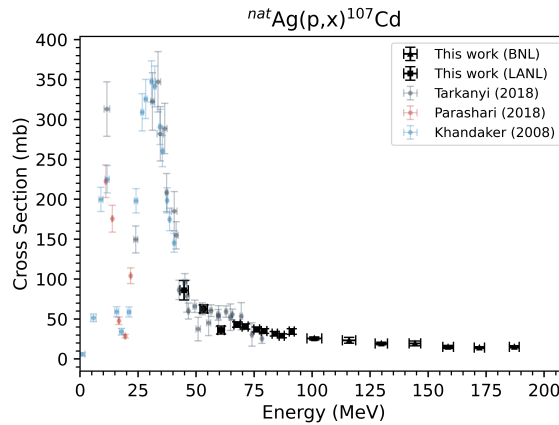


Figure 2.10: Independent cross section measurements of ${}^{\text{nat}}\text{Ag}(\text{p},\text{x}){}^{107}\text{Cd}$ [21, 22, 23].

$^{\text{nat}}\text{Ag}(\text{p},\text{x})^{104}\text{Cd}_{(i)}$

Similar to the case of ^{107}Cd production, independent cross section measurements of $^{\text{nat}}\text{Ag}(\text{p},\text{x})^{104}\text{Cd}$ are reported and shown in Figure 2.11.

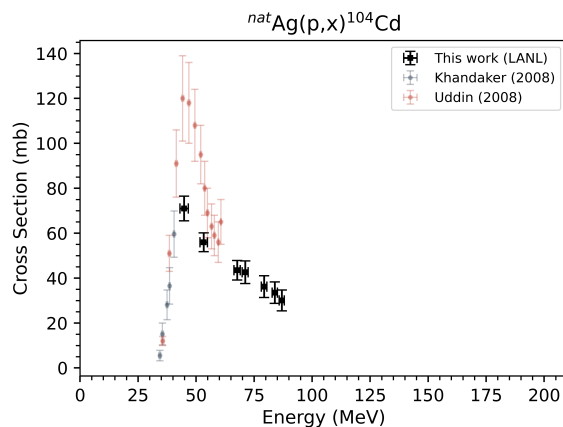


Figure 2.11: Independent cross section measurements of $^{\text{nat}}\text{Ag}(\text{p},\text{x})^{107}\text{Cd}$ [23, 24].

$^{\text{nat}}\text{Ag}(\text{p},\text{x})^{102}\text{Rh}_{(i)}$

Directly produced via $(\text{p},\text{p}\alpha\text{xn})$ reactions, the independent cross sections for ^{102}Rh production were measured and presented in Figure 2.12. Independent cross section measurements involving outgoing alpha allows for the exploration of alpha optical potentials in reaction modeling.

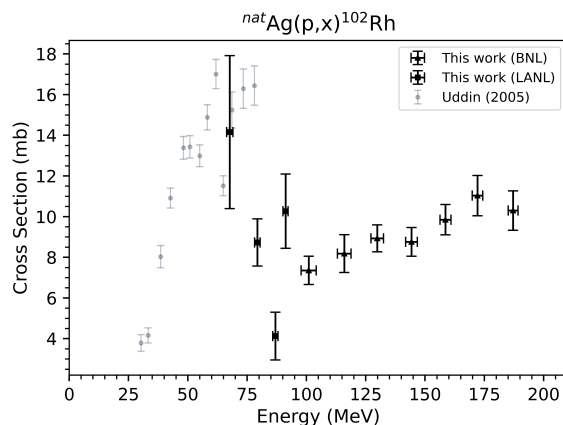


Figure 2.12: Independent cross section measurements of $^{\text{nat}}\text{Ag}(\text{p},\text{x})^{102}\text{Rh}$ [24].

$^{\text{nat}}\text{Ag}(\text{p},\text{x})^{101\text{m}}\text{Rh}_{(i)}$

With the decay feeding from ^{101}Pd accounted for, independent cross sections for $^{101\text{m}}\text{Rh}$ production were able to be measured (Figure 2.13). This adds to the dataset available for tuning the alpha optical potential in reaction modeling.

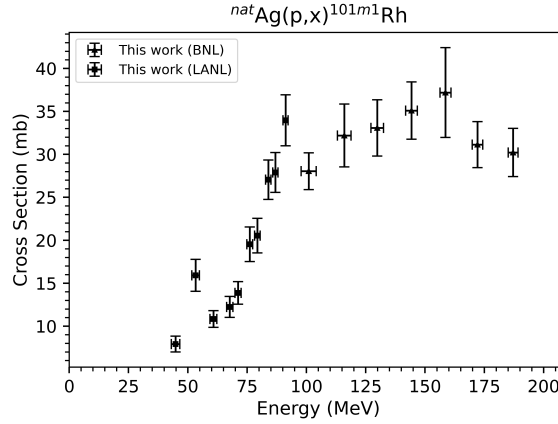


Figure 2.13: Independent cross section measurements of $^{\text{nat}}\text{Ag}(\text{p},\text{x})^{101\text{m}}\text{Rh}$.

$^{\text{nat}}\text{Ag}(\text{p},\text{x})^{100\text{m}+\text{g}}\text{Rh}_{(m+)}$

The total, isomer and ground-state, production cross section for ^{100}Rh were measured (Figure 2.14). While the decay feeding from ^{101}Pd was deconvolved, the decay of the short-lived ($t_{1/2} = 4.6(2)$ min) ^{100}Rh isomer was not. While discrepancy in magnitude with the measurement of Uddin *et al.* [12] is seen, the decay feeding from ^{101}Pd is included in Uddin *et al.*. Given the difference in measurement for ^{101}Pd production between Uddin *et al.* and this work, this discrepancy here is expected.

2.4.2 Evidence of Secondary Particle-Induced Reactions

Secondary particles were created primarily through reactions of the high-energy incident protons and thick degrader material ($^{\text{nat}}\text{Cu}$). Secondary charged particles are expected to be mostly stopped in the thick degraders, yet secondary neutrons could traverse the rest of the stack. The effects of secondary-neutron-induced reactions in stack-target experiments at BNL BLIP have been discussed in Apgar *et al.* [6] and can be traced back to cross section measurements reported in Fox *et al.* [4]. The increase of neutron flux towards the low-energy end of the stack, compounded by neutron moderation in surrounding cooling water, result in substantial contamination to cross section measurements for isotopes that can be simultaneously produced via (n,x) reactions. In this work, such effects are also clearly seen; selected examples are discussed below. Full characterization of the secondary particle

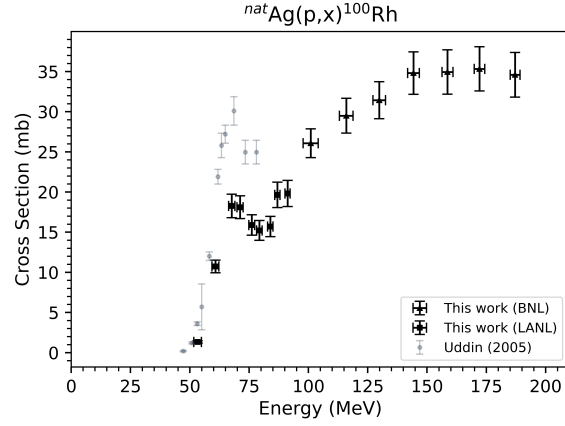


Figure 2.14: Total cross section measurements of $^{\text{nat}}\text{Ag}(p,x)^{100}\text{Rh}$ [24].

spectra is out of the scope of this work. However, the presence of co-production of these radionuclide products causes them to be excluded from use for optimizing the reaction model performance.

$^{\text{nat}}\text{Ag}(p,x)^{106\text{m}}\text{Ag}_{(i)}$

As seen in Figure 2.15, there is a jump in excitation function between the lowest energy data point from the BNL stack (101.0(32) keV) and the highest energy data point from the LANL stack (91.2(11) keV). $^{106\text{m}}\text{Ag}$ can be directly produced via both $^{\text{nat}}\text{Ag}(p,pxn)$ and $^{\text{nat}}\text{Ag}(n,xn)$ reactions.

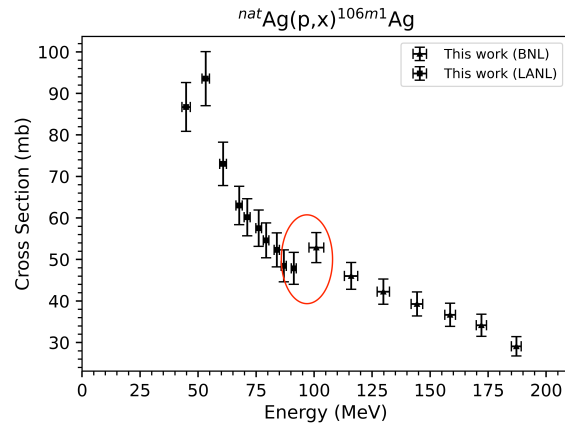


Figure 2.15: Independent cross section measurements of $^{\text{nat}}\text{Ag}(p,x)^{106\text{m}}\text{Ag}$. The discontinuity in excitation function is circled in red.

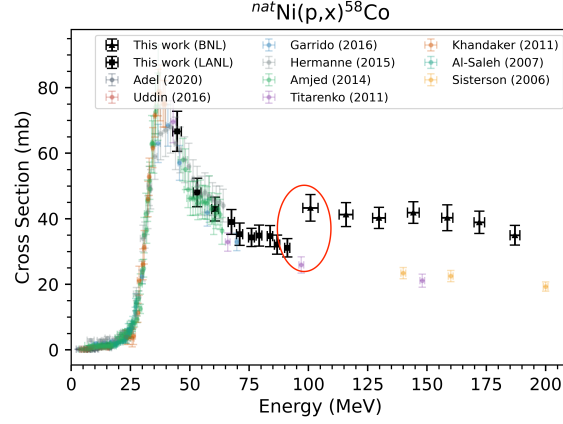


Figure 2.16: Total cross section measurements of ${}^{\text{nat}}\text{Ni}(p,x){}^{58}\text{Co}$ [25, 26, 27, 28, 29, 30, 31, 32, 33]. The discontinuity in excitation function is circled in red.

${}^{\text{nat}}\text{Ni}(p,x){}^{58}\text{Co}_{(m+)}, {}^{57}\text{Ni}_{(c)}$

Initially chosen as potential monitor reactions for beam characterization, especially for the BNL stack, ${}^{\text{nat}}\text{Ni}(p,x){}^{58}\text{Co}$ and ${}^{\text{nat}}\text{Ni}(p,x){}^{57}\text{Ni}$ were ultimately excluded due to concerns of co-production via secondary-neutron-induced reactions. As expected, jumps in the cross section measurements between the lowest energy data point from the BNL stack and the highest energy data point from the LANL stack were observed (Figure 2.16, 2.17).

For reactions on lower-Z nuclei such as those of ${}_{28}\text{Ni}$, possibilities of (n,pxn) reactions could be as significant as (n,xn) reactions. Such reactions lead to the co-production of Co isotopes, as shown in the example of ${}^{\text{nat}}\text{Ni}(p,x){}^{58}\text{Co}$.

In both cases, the agreement with literature data for measurements of proton energies lower than 100 MeV (from the LANL stack) yet significant deviations for proton energies higher than 100 MeV (from the BNL stack) further supports the occurrence of secondary-neutron induced reactions.

2.5 Charged-Particle Reaction Modeling with Talys-2.0

The system of interactions among nucleons in a reaction satisfies the non-relativistic time-independent radial Schrodinger Equation [15],

$$-\frac{\hbar^2}{2m} \left(\frac{d^2 R}{dr^2} + \frac{2}{r} \frac{dR}{dr} \right) + \left[\mathcal{U}(r) + \frac{\ell(\ell+1)\hbar^2}{2mr^2} \right] R = ER \quad (2.6)$$

, where m is the incident particle mass, R is the radial part of the wave function, E is the energy eigenvalue, and $\mathcal{U}$ is the potential well. The essence of Equation 2.6 is captured in

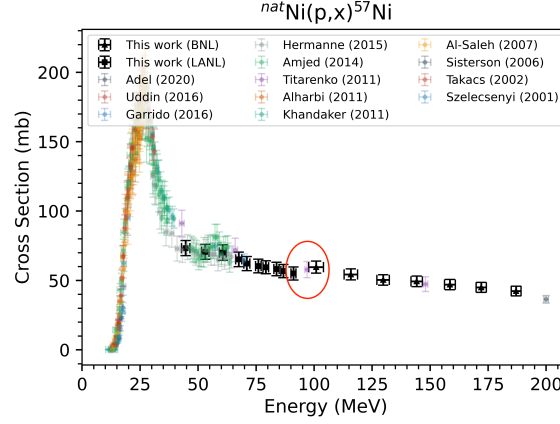


Figure 2.17: Cumulative cross section measurements of $^{nat}\text{Ni}(p,x)^{57}\text{Ni}$ [25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36]. The discontinuity in excitation function is circled in red.

Talys [37], one of the many prominent reaction modeling software packages available. With its outstanding user-friendliness in accessibility to underlying models and faster computational calculations with deterministic methods, Talys has been chosen over other options for all past TREND works [3, 4, 5, 6, 7]. Of particular relevance, in Apgar *et al.*, [6], the optimization of Talys parameters was informed by cross section measurements of $^{nat}\text{Sb}(p,x)$ reactions; with the proximity of $^{nat}_{51}\text{Sb}$ and $^{nat}_{47}\text{Ag}$ isotopes on the chart of the nuclides, optimized parameters from Apgar *et al.* serve as a helpful starting point for this iteration of the Talys parameter exploration in TREND. It should be noted that proton energies cover from approximately 40 MeV up to 200 MeV only in this work, as opposed to the threshold to 200 MeV energy range of most past TREND works. This resulted in few observed compound peaks in the measured excitation functions, and hence generally lower cross section values for Talys parameter optimization.

2.5.1 Summary of Reaction Modeling in Talys

Suitable for incident particle energies up to 200 MeV, Talys starts the calculation of every reaction with the modeling of competitions between compound, pre-equilibrium, and direct reaction mechanisms, as shown in Figure 2.18. In short, the higher the incident particle energy, the less equilibrated the compound nucleus is before subsequent particle emissions, if formed at all. This, however, is only the first step in modeling process, since the emissions of residual nuclei decay have to be accounted for. At higher energies, multiple particle emissions can occur; for every residual nucleus, Hauser-Feshbach calculations involving optical models, level densities, and photon strength functions are performed before the reaction is concluded. An overview of the process is provided below. Fission is ignored for these low- Z nuclei under consideration.

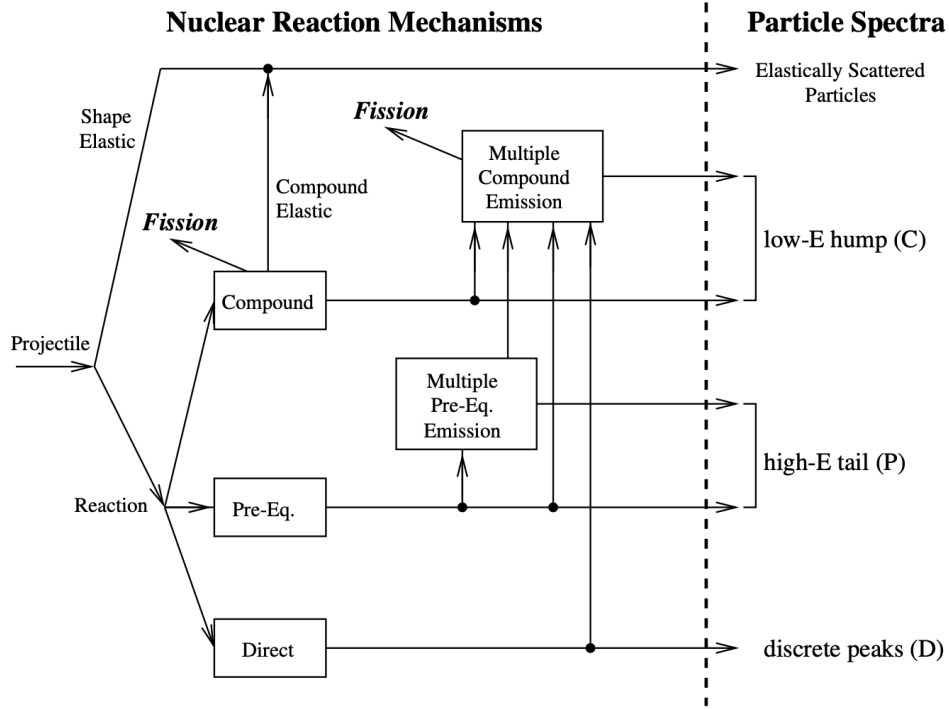


Figure 2.18: Competing reaction mechanisms, taken from the Talys manual [37].

Hauser-Feshbach Calculations

In general, cross section calculations can be formulated either by the scattering/absorption of incident particles in an averaged potential well created by the target nucleons, or the explicit tracking of reaction procession through a series of resonances.

By adopting the optical model, it is assumed that the reactions between the incident particle and the massive target nucleus can be described by a complex mean potential. Overall, the optical model yields a total cross section for a given orbital angular momentum ℓ of [38]:

$$\langle \sigma_\ell(E) \rangle_{\text{optical}} = \pi \lambda^2 (2\ell + 1) T_\ell \quad (2.7)$$

, where λ is the reduced wavelength, and T_ℓ is the calculated transmission coefficient.

In the case where reactions are described by a series of resonant scatterings through well-separated resonances, reaction $X(a,b)Y$ can be described by [38]:

$$\sigma_{ab}^J(E) = \pi \lambda^2 \omega_a^J \sum_{\lambda} \frac{\Gamma_{\lambda a} \Gamma_{\lambda b}}{(E - E_\lambda)^2 + \Gamma_\lambda^2/4} \quad (2.8)$$

, where J is the spin, E is the incident particle energy, $\Gamma_{\lambda a}$ and $\Gamma_{\lambda b}$ are partial widths of the Breit-Wigner resonance, Γ_γ is the total width, E_γ is the energy of the resonance, and ω_a^J is

a statistical factor given by equation 2.9.

$$\omega_a^J = \frac{2J+1}{(2i_a+1)(2I_a+1)} \quad (2.9)$$

, where i_a and I_a are the spins of the projectile and target respectively.

Integrating over an energy interval containing many ($\gg 1$) resonances with average spacing D , the energy-averaged cross section is:

$$\langle \sigma_{ab}^J(E) \rangle = \pi \lambda^2 \omega_a^J \frac{2\pi}{D} \left\langle \frac{\Gamma_a \Gamma_b}{\Gamma} \right\rangle \quad (2.10)$$

Defining a width fluctuation factor W_{ab} , the energy-averaged cross section can be further written as:

$$\langle \sigma_{ab}^J(E) \rangle = \pi \lambda^2 \omega_a^J \frac{2\pi}{D} W_{ab} \frac{\langle \Gamma_a \rangle \langle \Gamma_b \rangle}{\langle \Gamma \rangle} \quad (2.11)$$

Summing over possible outgoing channels b for the total reaction cross section,

$$\sigma_a^J(E) = \sum_b \sigma_{ab}^J(E) = \pi \lambda^2 \omega_a^J \sum_\lambda \frac{\Gamma_{\lambda a} \Gamma_\lambda}{(E - E_\lambda)^2 + \Gamma_\lambda^2/4} \quad (2.12)$$

, and similarly the energy-averaged cross section will be given by:

$$\langle \sigma_a^J(E) \rangle = \pi \lambda^2 \omega_a^J \frac{2\pi \langle \Gamma_a \rangle}{D} \quad (2.13)$$

In the simplified picture where the projectile and target are spinless and hence $\omega_a^J = 2\ell + 1$, the cross section becomes:

$$\langle \sigma_\ell(E) \rangle = \pi \lambda^2 (2\ell + 1) \frac{2\pi \langle \Gamma_\ell \rangle}{D} \quad (2.14)$$

Comparing to the results of the optical model given in Equation 2.7, it can be seen that the transmission coefficient can now be expressed as:

$$T_\ell = \frac{2\pi \langle \Gamma_\ell \rangle}{D} \quad (2.15)$$

Finally, by incorporating angular momentum and parity conservation, the Hauser-Feshbach expression for energy-differential cross sections is given by:

$$\frac{d\sigma_{ab}^{HF}}{dE_b} = \pi \lambda \sum_{J\Pi} \omega_a^J \frac{\sum_{\{j_a\}} T_a}{\sum_c \sum_{\{n_c\}} \sum_{\{j_c\}} T_c} \sum_{I_b \Pi_b} \sum_{\{j_b\}} T_b \rho(E_b, I_b, \Pi_b) \quad (2.16)$$

, where $\{j_i\}$ represents the set of angular momenta, $\{n_i\}$ is the set of states, Π is the parity of the compound system, and $\rho(E_b, I_b, \Pi_b)$ is the level density of the final states.

Reaction mechanisms and models for residual nuclei decay relevant to this work are discussed in the following subsections.

2.5.2 Fitting Procedure of Talys-2.0 Parameters

Parameter adjustments were based on independent or total cross sections of six reactions including $^{\text{nat}}\text{Ag}(\text{p},\text{x})^{107,104}\text{Cd}$, $^{106\text{m}}\text{Ag}$, $^{102,101\text{m},100}\text{Rh}$. Data extracted from the BNL stack for $^{\text{nat}}\text{Ag}(\text{p},\text{x})^{106\text{m}}\text{Ag}$ were excluded from the analysis due to the clear contamination from secondary-particle-induced reactions.

Validation was performed using the cumulative cross sections of $^{103,101,100}\text{Pd}$, ^{97}Rh , ^{97}Ru , and ^{96}Tc production. Since the modeling of cumulative channels is complicated by the convoluted calculations of parent nuclei production cross sections, no cumulative channels were used in the optimization process. They however still served to validate the improvement in parameter adjustments. The $^{103,101,100}\text{Pd}$ channels were chosen due to their relevance to the motivation of this work and the higher relative cross section magnitudes. The ^{97}Rh , ^{97}Ru , and ^{96}Tc production channels were chosen to ensure the inclusion of potential α -particle emission calculations. No Ag isotope production channels were used for validation due to concerns of the aforementioned impact of secondary-particle-induced reactions.

Despite the wide availability of more than 400 tunable parameters in Talys, only a subset of those of relevance and with physically-justifiable changes are explored.

The fitting procedure is as follows [7]:

1. Selection of independently-measured reaction cross section data for “training”.
2. Determination of goodness-of-fit metric.
3. Optimization of base-model selection parameters, including those of level density, photon strength, and α -particle optical model, *e.g.*, `ldmmodel`, `strength`, and `alphaomp`.
4. Adjustments of optical model parameters, *e.g.*, `w1adjust` and `w2adjust` for protons and neutrons.
5. Adjustments of pre-equilibrium parameters, *e.g.*, `M2constant`, `M2limit`, and `Rpinu`.
6. Iterations of steps 4 and 5 until convergence in the goodness-of-fit metric.
7. Validation with selected cumulative reaction cross section data.

The goodness-of-fit metric chosen for this work is based on past similar TREND works [3, 4, 5, 6, 7]. The total goodness-of-fit across all utilized channels is calculated as:

$$\chi_{\text{tot}}^2 = \sum_{c=1}^{N_c} \chi_c^2 w_c \quad (2.17)$$

, where N_c is the number of channels in consideration and w_c is the weighting factor for the channel. χ_c^2 , the goodness-of-fit for each channel c , is determined by:

$$\chi_c^2 = \frac{1}{N_p} \sum_{i=1}^{N_p} \left(\frac{\sigma_T^i - \sigma_E^i}{\Delta\sigma_E^i} \right)^2 \quad (2.18)$$

, where N_p is the number of data points used for the channel, σ_T is the Talys-calculated cross section, and σ_E is the experimentally-measured cross section with $\Delta\sigma_E$ uncertainty. To account for the difference in prominence of each channel, the weighting factor for each channel is calculated as:

$$w_c = \frac{\sum_{i=1}^{N_p} \sigma_E^{ci}}{\sum_{e=1}^{N_e} \sum_{i=1}^{N_p} \sigma_E^{ci}} \quad (2.19)$$

It should be noted that uncertainties in proton energies were not taken into account during the optimization process.

Available options and parameters of explored models in Talys-2.0 are discussed in the following sections.

Table 2.1: Summary of Talys-2.0 base-model parameter exploration [37].

Parameter	Best	Default	Options
<code>ldmodel</code>	6	1	1-6
<code>strength</code>	8	9	1-10
<code>deuteronomp</code>	1	1	1-5
<code>alphaomp</code>	3	6	1-8
<code>preeqmode</code>	2	2	1-4
<code>preeqspin</code>	1	1	1-3

2.5.3 Level Density and Photon Strength in Talys-2.0

Included in the base-models of Talys-2.0 (see Table 2.1), level density and photon strength functions are part of the first set of parameters for optimization.

Level density models are invoked at excitation energies where there is no available discrete level data, as in Equation 2.16. In Talys, both phenomenological and microscopic models are available [37]. For this work, the microscopic model based on temperature-dependent Gogny-Hartree-Fock-Bogolyubov (HFB) calculations (`ldmodel` 6) outperforms all other options [39] by times ranging from 1.19 to 2.68 in the goodness-of-fit metric, as shown in Table 2.2. Phenomenological options include the Constant Temperature + Fermi Gas Model (`ldmodel` 1), the Back-shifted Fermi gas Model (`ldmodel` 2), and the Generalized Superfluid Model (`ldmodel` 3) [37]. Other available microscopic models (`ldmodel` 4-5) are based on the Skyrme-HFB calculations [37].

The photon strength function governs the de-excitation of nuclei via photon emission and is important in determining the population of specific low-lying states in final residual products. Photon transmission coefficients that are directly proportional to photon strength functions are incorporated into Hauser-Feshbach calculations, modeling the competition between photon and particle emission [37]. Similar to the case of level density models, Talys makes available both phenomenological and microscopic photon strength modeling options to

Table 2.2: Level density models in Talys-2.0.

ldmodel	χ_{tot}^2
1	28.51
2	21.42
3	24.42
4	18.68
5	42.13
6	15.69

the user. Options **strength 1,2,5** adopt various analytical descriptions of the giant dipole resonance shape with or without particular corrections for E1 transitions [37]. Microscopic options include QRPA corrected Skryme-HFB models, etc. The Gogny-HFB model with QRPA (**strength 8**) outperformed all other options, as preferred by the authors of Talys [37, 40].

In both cases of the level density and photon strength function, HFB calculations incorporating the D1M version of the Gogny force create the best agreement with experimental data in this work.

2.5.4 Optical Model in Talys-2.0

The exploration of optical model parameters in Talys follows the work of Apgar *et al.* [6] and Morrell *et al.* [7]. The formulation of optical model potentials (OMP) in Talys is summarized as follows [37].

The general OMP, which consists of real $\mathcal{V}$ and imaginary $\mathcal{W}$ components of the volume-central V , surface-central D , spin-orbit SO , and Coulomb C potentials, is described as:

$$\begin{aligned} \mathcal{U}(r, E) = & -\mathcal{V}_V(r, E) - i\mathcal{W}_V(r, E) - i\mathcal{W}_D(r, E) \\ & + \mathcal{V}_{SO}(r, E) \cdot \ell \cdot \sigma + i\mathcal{W}_{SO}(r, E) \cdot \ell \cdot \sigma + V_C(r) \end{aligned} \quad (2.20)$$

Each V , D , and SO potential is characterized by an energy-dependent well depth and a radially dependent Woods-Saxon shaped form factor, as shown in Equation 2.21.

$$\begin{aligned}
 \mathcal{V}_V(r, E) &= V_V(E)f(r, R_V, a_V), \\
 \mathcal{W}_V(r, E) &= W_V(E)f(r, R_V, a_V), \\
 \mathcal{W}_D(r, E) &= -4a_D W_D(E) \frac{d}{dr} f(r, R_D, a_D), \\
 \mathcal{V}_{SO}(r, E) &= V_{SO}(E) \left(\frac{\hbar^2}{m_\pi c} \right)^2 f(r, R_{SO}, a_{SO}), \\
 \mathcal{W}_{SO}(r, E) &= V_{SO}(E) \left(\frac{\hbar^2}{m_\pi c} \right)^2 f(r, R_{SO}, a_{SO})
 \end{aligned} \tag{2.21}$$

The C potential for uniformly-charged spherical nucleons is calculated using Equation 2.22:

$$\begin{aligned}
 \mathcal{V}_C(r) &= \frac{Zze^2}{2R_C} \left(3 - \frac{r^2}{R_C^2} \right), \text{ for } r \leq R_c \\
 &= \frac{Zze^2}{r}, \text{ for } r \geq R_c
 \end{aligned} \tag{2.22}$$

, where $R_c = r_C A^{1/3}$.

Following the work of Apgar *et al.* [6], this exploration of Talys OMP parameters focuses on the adjustments to the imaginary volume-centric potential $\mathcal{W}_V$, where its energy-dependent well depth can be further parameterized as in Equation 2.23.

$$\begin{aligned}
 W_V(E) &= \mathbf{w1} \frac{(E - E_f)^2}{(E - E_f)^2 + (\mathbf{w2})^2} \\
 \mathbf{r_V} &= \text{constant} \\
 \mathbf{a_V} &= \text{constant}
 \end{aligned} \tag{2.23}$$

, where E_f is the Fermi energy, and $\mathbf{w1}$, $\mathbf{w2}$, $\mathbf{r_V}$, and $\mathbf{a_V}$ are adjustable with keywords **w1adjust**, **w2adjust**, **rvadjust**, and **avadjust**, respectively. Since the terms $\mathbf{r_V}$ and $\mathbf{a_V}$ build the geometry of the Wood-Saxon form factor, adjustments to **rvadjust** and **avadjust** also affect the real volume-centric potential $\mathcal{V}_V$. These keywords are hence excluded from the parameter optimization process, since changes to the nuclear radius are difficult to justify.

For **w1adjust** and **w2adjust** of protons and neutrons, imposed constraints of 0.1-4.0 were applied. The resulting optimized OMP parameters are shown in Table 2.3. Similar to the results of Morrell *et al.* [7], optimized **w1adjust** for both protons and neutrons fell at the boundary of constraints, increasing the imaginary well depth. The relatively large adjustments to **w1adjust** p and n result in significant increases of the non-elastic cross sections of higher energies.

In addition to the OMP of protons and neutrons, complex particle OMPs are also adjustable in Talys. With independent cross section measurements of $^{102,101m,100}\text{Rh}$ production, the alpha OMP and deuteron OMP models were also investigated with keywords **alphaomp**

Table 2.3: Summary of Talys-2.0 optical model parameter exploration [37]. Parameters optimized at the limit of the constraints are marked with a *.

Parameter	Best	Constraints	Default	Range
w1adjust n	4.0*	0.1-4.0	1.0	0.1-10.0
w2adjust n	1.5	0.1-4.0	1.0	0.1-10.0
w1adjust p	4.0*	0.1-4.0	1.0	0.1-10.0
w2adjust p	2.6	0.1-4.0	1.0	0.1-10.0

and **deuteronomp**, respectively. The resulting “best” options and default options are included in Table 2.1. **alphaomp 3** adopts the double-folding method of Demetriou *et al.* [41], and **deuteronomp 1** utilizes a simplification of the folding techniques of Watanabe [42].

2.5.5 Pre-Equilibrium in Talys-2.0

Details of the exciton model for pre-equilibrium reactions in Talys can be found in the manual [37] and Fox *et al.* [3]. Here, only a summary with an explanation of adjusted parameters is presented.

In an exciton model, the scattering process is characterized by the progression through various configurations of the number of particles above and holes below the Fermi surface, which are indiscriminately referred to as excitons [37]. In the two-component exciton model of Talys, the transition rates of the production, annihilation, and conversion of particle-hole pairs determined by Fermi’s golden rule are expressed in terms of effective squared matrix elements [37]. For instance, the collision probability per unit time of a proton-neutron interaction leading to the conversion of a proton particle-hole pair to a neutron particle-hole pair is written as

$$\lambda_{\pi\nu}^{1p1h} = \frac{2\pi}{\hbar} M_{\pi\nu}^2 \omega \quad (2.24)$$

, where ω is the particle-hole state density, dependent on the exciton configuration. The two-component matrix elements are further parameterized as shown in Equation 2.25.

$$\begin{aligned} M_{\pi\pi}^2 &= R_{\pi\pi}^2 M^2 \\ M_{\pi\nu}^2 &= R_{\pi\nu}^2 M^2 \\ M_{\nu\pi}^2 &= R_{\nu\pi}^2 M^2 \\ M_{\nu\nu}^2 &= R_{\nu\nu}^2 M^2 \end{aligned} \quad (2.25)$$

In Talys, the semi-empirical averaged squared matrix element is calculated as:

$$M^2 = \frac{C_1 A_p}{A^3} \left\{ 7.48 C_2 + \frac{4.62 \times 10^5}{[E^{\text{tot}}/(n \cdot A_p) + 10.7 C_3]^3} \right\} \quad (2.26)$$

, where n is the total exciton number and A_p is the mass number of the projectile. Constants C_1 , C_2 , and C_3 are accessible via keywords **M2constant**, **M2limit**, and **M2shift** respectively. Accounting for the difference in interactions of like or unlike nucleons, terms R_{xy} , $x = \pi, \nu$ in Equation 2.25 are also adjustable through keywords **Rpipi**, **Rpinu**, **Rnupi**, and **Rnunu**.

The outcome of pre-equilibrium parameters optimization is shown in Table 2.4. The most noticeable adjustments performed were the enhancement of the neutron-proton ratio of the matrix element and the suppression of the proton-neutron ratio and the proton-proton ratio.

Table 2.4: Summary of Talys-2.0 pre-equilibrium model parameter exploration [37]. Parameters optimized at the limit of the constraints are marked with a *.

Parameter	Best	Constraints	Default	Range
M2constant	1.2	0.1-5.0	1.0	0.0-100.0
M2shift	1.0	0.1-5.0	1.0	0.0-100.0
M2limit	1.0	0.1-5.0	1.0	0.0-100.0
Rnunu	1.3	0.1-5.0	1.5	0.0-100.0
Rnupi	2.0	0.1-5.0	1.0	0.0-100.0
Rpinu	0.3	0.1-5.0	1.0	0.0-100.0
Rpipi	0.1*	0.1-5.0	1.0	0.0-100.0

Other pre-equilibrium reaction related parameters include **preeqmode** and **preeqspin** shown in Table 2.1. **preeqmode** 2 invokes exciton model calculations with numerical transition rates and energy-dependent matrices; **preeqspin** 1 adopts the compound nucleus spin distribution for residual states after pre-equilibrium emissions.

2.5.6 Comparisons of the Modeled and the Measured

Table 2.5: Summary of Talys-2.0 modeling results.

	Training χ_{tot}^2	Validation χ_{tot}^2
Optimized Parameters	8.73	19.94
Default Parameters	28.51	28.45
Apgar Parameters [6]	20.18	22.13
Morrell Parameters [7]	34.26	26.87

Adjustments to the Talys-2.0 parameters resulted in significant improvements in predictive capabilities of cross sections, as shown in Table 2.5. Improvement ratios of 3.27 and 1.45 for the χ_{tot}^2 figure-of-merit were observed over default for the training and validation channels respectively. The goodness-of-fit for each channel, χ_c^2 , is presented in Table 2.6. Results of

adjustments performed using optimized parameters of Apgar *et al.* [6] and Morrell *et al.* [7] are also provided for reference. Figure 2.19 shows the comparison of measured cross sections with Talys-2.0 predictions using the optimized parameters of this work, default values, those of Apgar *et al.* [6], and those of Morrell *et al.* [7]. While the Apgar parameters performed better than default as foreseen, since only (p,xn) reactions were used in the optimization process, they were not able to predict production cross sections of Rh isotopes as well as they do for Cd and Ag isotopes. On the other hand, the Morrell parameters performed noticeably better than the Apgar parameters in predictions of production cross sections of Rh isotopes, yet fails to produce the same level of accuracy for Cd and Ag isotopes. The Morrell parameters were informed partly by (p,pxn), (p,dxn), and (p,axn) reactions, hence likely yielding OMP parameter adjustments that closer align with those of this work. As expected, improvements for the goodness-of-fit of validation channels were not as pronounced as seen in Figure 2.20. While for certain channels, the Apgar, Morrell, or even default parameters outperform the adjusted parameters, an overall improvement was still observed.

Table 2.6: χ_c^2 for individual training and validation channels.

	χ_c^2 Optimized	χ_c^2 Default	χ_c^2 Apgar	χ_c^2 Morrell
^{107}Cd	4.50	20.21	7.80	24.66
$^{106\text{m}}\text{Ag}$	3.62	10.42	3.41	67.91
^{104}Cd	1.02	9.17	4.15	9.55
^{102}Rh	14.09	86.28	64.634	58.15
$^{101\text{m}}\text{Rh}$	27.36	65.22	60.47	35.78
^{100}Rh	8.69	37.61	28.71	5.38
^{103}Pd	14.82	13.49	13.18	10.39
^{101}Pd	13.91	28.48	15.31	31.37
^{100}Pd	38.32	77.64	48.62	76.08
^{97}Rh	94.95	382.74	286.69	349.95
^{97}Ru	98.03	111.88	90.25	149.18
^{96}Tc	5.95	114.35	83.49	56.23

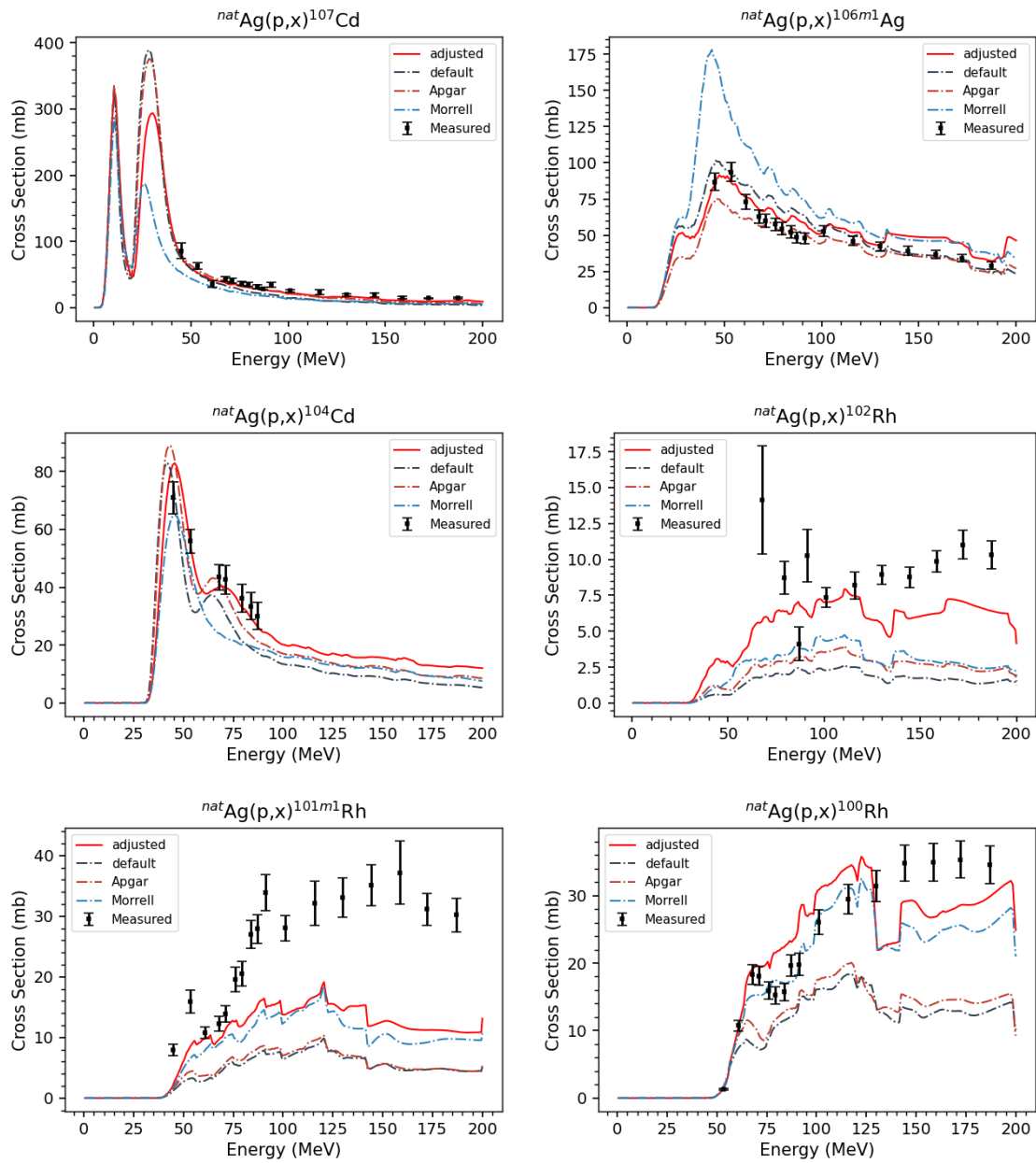


Figure 2.19: Comparison of modeled and measured cross section values for reaction channels used for Talys-2.0 parameter training.

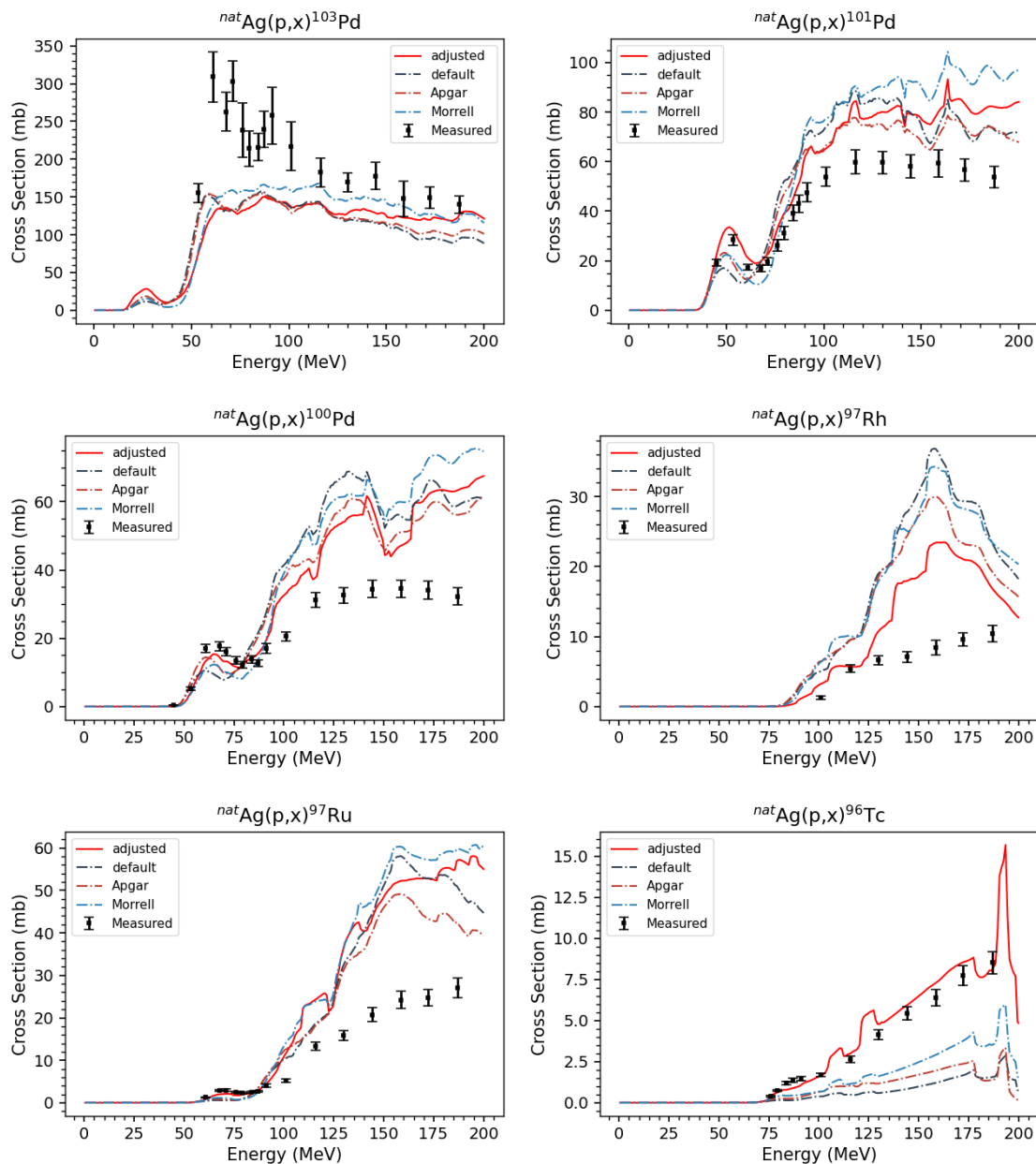


Figure 2.20: Comparison of modeled and measured cross section values for reaction channels used for Talys-2.0 parameter validation.

2.5.7 Further Investigation of the Level Density Model

As seen in Table 2.6, the fit of the modeled to the measured $^{\text{nat}}\text{Ag}(p,x)^{101m}\text{Rh}$ excitation function was not as successful as for the other channels used for parameter training. It is suspected that this could be caused by the current lack of understanding in spin distributions of levels in the quasi-continuum. Talys allows for an adjustment of the spin cut-off parameter, **Rspincut**, which scales the width of the angular momentum distribution of level densities [37]. Thoroughly investigated in Apgar *et al.* [6], an adjustment to 0.4 from the default value of 1.0 was concluded.

The spin cut-off parameter is only in effect when a phenomenological level density model is invoked, making this change impossible for the chosen optimized parameters since a microscopic level density model was utilized. A brief attempt was made to explore the potential impact of **Rspincut** adjustments on the ^{101m}Rh production channel by temporarily adopting the best phenomenological model, namely the Back-shifted Fermi Gas Model (**ldmodel 2**). All other parameters (photon strength, OMP, pre-equilibrium) were set to default. Talys-2.0 allows for **Rspincut** values of 0.0-10.0, but only range 0.1-2.0 was explored in increments of 0.1. As shown in Figure 2.21, the χ_c^2 -minimizing value of **Rspincut** is 0.8 for this reaction. Since only one reaction with low cross section values was used to inform this process, this adjustment is **in no way** as compelling as what was presented in Apgar *et al.* [6], where multiple production channels of isomer-ground state pairs of the same isotope with well-characterized compound peaks were used for parameter training. It is, however, indicative of the need for further studies in the distribution of spins and population of states in the quasi-continuum of nuclear levels.

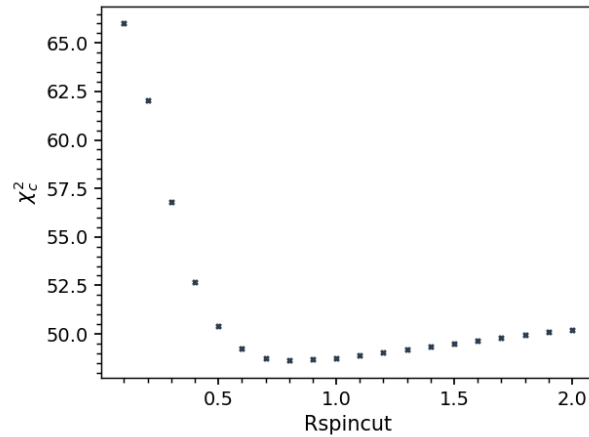


Figure 2.21: χ_c^2 for $^{\text{nat}}\text{Ag}(p,x)^{101m}\text{Rh}$ with varying **Rspincut**.

2.6 Conclusions and Future Considerations

A stacked-target irradiation was each conducted at the LANL IPF and BNL BLIP, generating cross section data for 24 $^{nat}\text{Ag}(\text{p},\text{x})$ channels in between 40 and 200 MeV, including first measurements of several channels. Additionally, 23 $^{nat}\text{Cu}(\text{p},\text{x})$ channels and 16 $^{nat}\text{Ni}(\text{p},\text{x})$ channels were characterized. A new set of cross sections of the $^{nat}\text{Ag}(\text{p},\text{x})^{103}\text{Pd}$ reaction were presented, along with first measurements of the reaction beyond 100 MeV, helping inform future designs of ^{103}Pd production routes at high-energy facilities.

Extending from past efforts in the TREND collaboration of Talys parameter explorations, an investigation in the modeling of level densities, photon strengths, optical model potentials, and pre-equilibrium reactions was conducted. While due to the decay properties of the isotopes produced and the fact that no low-energy measurement below 40 MeV was made, the number of reactions applicable in the Talys parameter optimization process is noticeably lower than in past TREND works, the results overall corroborate past findings. However, it should be noted that covariance among measured cross sections were not accounted for the parameter optimization process in this work, and should be further explored in future efforts.

While the stacked-target/foil irradiation method is long established, a call for caution is made in the possible negligence of secondary-particle-induced reactions. Particularly at higher energies, secondary particles born in thick degrader materials may significantly increase the apparent reaction cross section values of both the target of interest and monitor foils. With the complete reliance on monitor foils to characterize beam degradation through the stack, there are undoubtedly covariances among reported cross sections that were not taken into consideration. Since the moderation of secondary neutrons can be largely facility-dependent, such effects have to be comprehensively studied should secondary-neutron-induced reactions be accurately accounted for. It should be noted, however, that for the isotope production community, such reactions could be beneficial in boosting the yields of isotopes of interest.

Chapter 3

Measurement and Modeling of Nuclear Quasi-Continuum Properties from $^{\text{nat}}\text{Tl}(\text{p},\text{x})$ at 30 and 50 MeV

3.1 Abstract

To meet the demand for $^{202\text{g}}\text{Pb}$ from the field of geochronology, the TREND (Tri-Laboratory Effort in Nuclear Data) collaboration between Lawrence Berkeley (LBNL), Los Alamos (LANL), and Brookhaven (BNL) National Laboratories set out to measure the production cross section for $^{202\text{g}}\text{Pb}$ via the $^{\text{nat}}\text{Tl}(\text{p},\text{x})$ reaction. The long half-life ($t_{1/2} = 52.5(28) \times 10^3$ y) and lack of decay γ -rays of the isotope made the stacked-target approach described in Chapter 2 unacceptable for determining the $^{202\text{g}}\text{Pb}$ production cross section. For this work, the collaboration pivoted to the use of in-beam γ -ray spectroscopy for the quantification of prompt γ -ray transitions in ^{202}Pb following the (p,xn) reactions, coupled with reaction modeling. Two natural thallium foils were irradiated at LBNL's 88-Inch Cyclotron, one at incident proton energy of 50 MeV and the other at 30 MeV. Stacked-target measurements were also performed by the collaboration to measure the production cross section of $^{202\text{m}}\text{Pb}$, which has a half-life of 3.54(2) hours and decays via the emission of several strong γ -rays. In this work, multiple prompt γ -ray transitions in lead isotopes of $198 \leq A \leq 203$ were quantified. Measured γ -ray production cross sections were incorporated in the exploration of level density theories in the reaction modeling code Talys-2.0. The outcome of parameter explorations of level density and the optical model potential in Talys calls to further examination of the interplay between the two models in theoretical cross section calculations. The combination of the prompt γ -ray measurements in ^{202}Pb , the stacked target $^{202\text{m}}\text{Pb}$ data, and the theoretical calculations of Talys allowed for the the first-ever measured production cross section of $^{202\text{g}}\text{Pb}$, that is 184 ± 31 mb at 50 MeV and 770 ± 180 mb at 30 MeV.

3.2 Introduction

Efforts in isotope production are not limited to the development of medical applications of radiation; they include maintaining the inventory of stable and long-lived isotopes. This includes that of ^{202g}Pb , which has been used for geochronology via thermal ionization mass spectroscopy techniques and has been proposed as a reference material for accelerator mass spectroscopy [43, 44]. The Pb-U Chronometer, *i.e.*, naturally-occurring ^{238}U and ^{235}U decay chains into ^{206}Pb and ^{207}Pb respectively, allow for accurate dating of events from mass extinctions to lunar formation. ^{202g}Pb serves in spikes for mass fractionation correction for Pb isotopic analyses to sub-nanogram quantities [44]. The $^{nat}\text{Tl}(p,x)$ reaction has been proposed as a method of production for ^{202g}Pb to meet anticipated needs.

As demonstrated in Chapter 2, the stacked-target method is efficient in measuring cross sections over a wide range of incident particle energy. However, as in all foil activation methods, conducting γ -ray counting post-irradiation naturally leads to the inability to quantify the production of isotopes of long half-lives or those with no measurable γ -ray in decay. This includes ^{202g}Pb , with its half-life of $52.5(28) \times 10^3$ years and the complete lack of any observed decay γ -ray.

To measure the production cross section for ^{202g}Pb , in-beam spectroscopy techniques were adopted in this experiment to allow direct observation of feeding into the ground state of the ^{202g}Pb nucleus. While the techniques are conventional in the general field of nuclear physics, they are extremely rare for the purpose of cross section measurements for isotope production.

This work is complemented by a stacked-target measurement also partially performed at the 88-Inch Cyclotron of LBNL (which I also participated in); the stacked-target data analysis is primarily performed by other members of the TREND collaboration, Dr. Jonathan Morrell and Dr. Wilson Lin of BNL, and will be presented in a separate publication. Selected cross section values and parameters used for reaction modeling were however adopted in this work and are made clear when and how they were incorporated.

3.3 Experimental Methods

3.3.1 Experimental Setup

Two natural thallium foils, prepared by the Stable Isotope Materials and Chemistry Group at Oak Ridge National Laboratory [45], were irradiated at 30 MeV and 50 MeV incident proton energy respectively in Cave 4C at the LBNL 88-Inch Cyclotron during August 13-16, 2022. Used to be where the Gammasphere array was hosted, this experiment was the first time beam was sent into Cave 4C after an over-a-decade hiatus on any experimental work performed at this beam line.

As shown in Figure 3.1, γ -ray emissions from the $^{nat}\text{Tl}(p,x)$ reactions were measured by a Clover HPGe detector and an ORTEC GEM p-type coaxial HPGe detector. During

the experiment, the Clover detector was positioned at various discrete angles, including 60° , 70° , 80° , and 90° , to allow for observations of angular-dependent properties of the pre-equilibrium emissions. Since the Clover detector is an assembly of four Ge crystals, the Compton-scattered γ -rays from one crystal can be photoelectrically absorbed in a nearby crystal. Higher photo-peak efficiency can hence be achieved through coincident timing in “add-back” mode, where the energies detected in the two crystals in the coincidence event are summed to recover the full energy of the γ -ray. The ORTEC GEM detector was kept at 90° at all times. A 0.8 cm-thick aluminum attenuator was attached to the front of the ORTEC GEM detector, and a 0.2 cm-thick copper attenuator was added to the front of the Clover detector. The distances between the detectors and the target were kept constant throughout the experiment, at 25 cm and 41.5 cm for the Clover detector and the ORTEC GEM detector, respectively. Both detectors were calibrated with a ^{152}Eu source (Eckert & Ziegler, Valencia, CA 91355 USA).



Figure 3.1: Experimental setup of this work. The arrow indicates the direction of the beam.

Mesytec MDPP-16 digital pulse-processing modules were used for data acquisition. The beam current was measured at the Faraday cup at the end of the beam line with a Brookhaven Instruments Corporation (BIC) Model 1000C Current Integrator every second. More details of the Mesytec data acquisition systems can be found in the publication of Gordon *et al.* on the GENESIS array at the 88-Inch Cyclotron [46], after which this setup was modeled.

While, as seen in Figure 3.1, an array of six EJ-309 liquid scintillators was also configured for the experiment, the analysis of information acquired from the scintillators is outside of the scope of this work.

3.3.2 Data Acquisition

Given the convoluted challenges of attempting a first-of-a-kind experiment in the isotope production field and technical difficulties in setting up detection systems at a long-unused location, problems arose in data acquisition that were not discovered until well after the experiment was concluded. They are presented here for transparency in communicating the systematic uncertainties and assumptions made in this work.

The BIC Model 1000C Current Integrator

The thallium foils used for this experiment were designed for use in the stacked-target experiments and therefore had areal densities of approximately 70 mg/cm^2 [45]. While the thickness of the foils were well-suited for the stacked-target experiments, which relied on significant yields of radioactive residual products for production quantification, to maintain a reasonable reaction rate for γ -ray detection during this experiment, the current of the incident proton beam had to be lowered to an approximate nominal current of 60 pA. The BIC Model 1000C Current Integrator guarantees accuracy in current reading to as low as $1/3$ of 2 nA (or 667 pA), the lowest full-scale current setting available; the nominal current readout for this experiment was approximately $1/30$ of 2 nA (or 67 pA). To account for this, in this work, actual current values were assumed to be a scalar multiple of what is readout from the current integrator. In other words, the actual current $I(t[s]) = I_{\text{nominal}}(t[s]) \times c$, and the actual integrated charge between times t_1 and t_2 is hence $Q(t_1, t_2) = Q_{\text{nominal}}(t_1, t_2) \times c$.

The Mesytec Data Acquisition System

In post-processing of the collected spectra, it was discovered that the “efficiency” at which the Mesytec data acquisition system, used to record the data from both the ORTEC GEM and the Clover detectors, operated could vary by over a factor of 8. The effect was corrected by the following means.

Under usual circumstances where a data acquisition system does not have varying “efficiency”, the counts N_c^γ of a prompt γ -ray in a spectrum should be given by:

$$N_c^\gamma = \sigma^\gamma \times (\rho R) \times Q \times \varepsilon_{\text{detection}}(E_\gamma) \quad (3.1)$$

, where σ^γ is the γ -ray production cross section, (ρR) is the areal density of the foil, Q is the integrated proton charge during the counting time interval, and $\varepsilon_{\text{detection}}(E_\gamma)$ is the efficiency of the detection system at energy E_γ . It was further noticed and therefore assumed that the change in data acquisition “efficiency” was likely occurring at every start of a new spectrum file, and not gradual over time. Assigning that the relative data acquisition “efficiency” is 1. in the first spectrum (*i.e.*, $\varepsilon_{\text{DAQ}, 0} = 1$.) of each run, for every other spectrum i ,

$$\varepsilon_{\text{DAQ}, i} = \frac{N_{c, i}^\gamma / Q_i}{N_{c, 0}^\gamma / Q_0} \quad (3.2)$$

, since σ^γ and $\varepsilon_{\text{detection}}(E_\gamma)$ do not vary from spectrum to spectrum for the same γ -ray at the same incident proton energy. The γ -ray chosen for this process for all spectra recorded with incident proton energy 50 MeV was the 1026.49 keV prompt transition in ^{200}Pb . For all spectra measured at 30 MeV, the 1014.1 keV prompt transition in ^{201}Pb was exploited. These transitions were chosen due to their high intensity, them being well-resolved (free from γ -ray peak multiplets), and their proximity to the 960.67 keV peak in ^{202}Pb , which was crucial for this work. To remove the angular dependence of σ^γ in the calculation of ε_{DAQ} , only the spectra collected from the ORTEC GEM detector were utilized for this purpose.

3.3.3 Data Interpretation

To quantify the production of ^{202g}Pb , measurements of partial γ -ray cross sections and theoretical calculations from the reaction-modeling code Talys were combined with cross section data obtained through a complementing stacked-target experiment. Through in-beam spectroscopy of this work and the measured production cross sections of ^{202m}Pb , the yrast $2^+ \rightarrow 0^+$ transition in ^{202}Pb was quantified. To account for ground state feeding from the quasi-continuum, theoretical models of Talys were employed. The measurements of γ -ray production cross sections are detailed in Section 3.4. The optimization of Talys parameters is discussed in Section 3.5. Finally, the calculation of the ^{202g}Pb production cross section is presented in Section 3.6. Figure 3.2 is a visual summary of the data interpretation performed in this work.

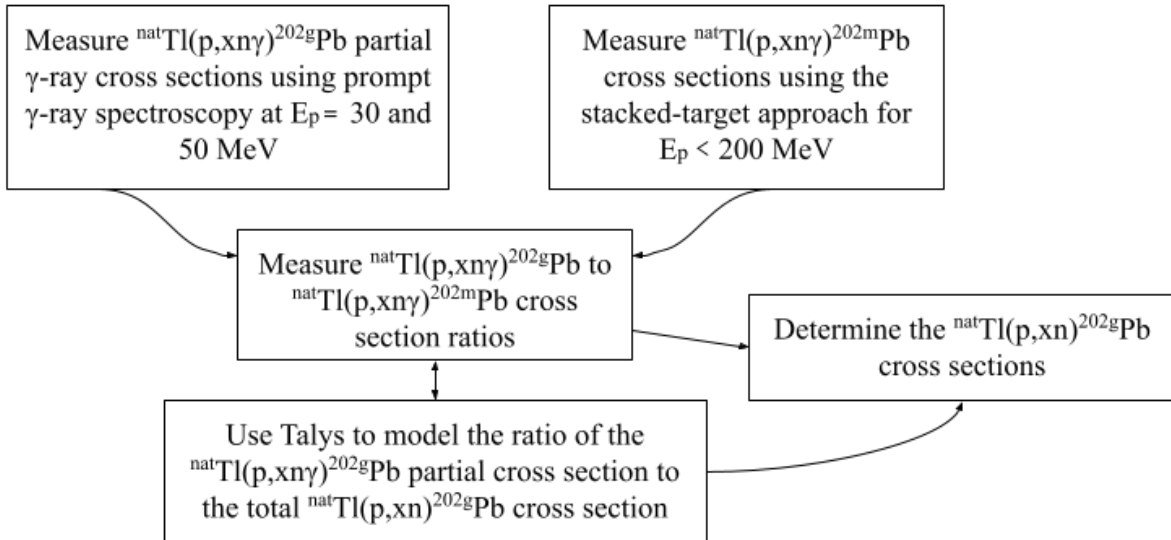


Figure 3.2: Summary of the data interpretation performed in this work.

3.4 Partial γ -Ray Cross Section Measurements

Partial γ -ray cross section measurements are central to this work, as they are what could only be achieved via in-beam spectroscopy. In this section, the derivation of the γ -ray production cross section of the the yrast $2^+ \rightarrow 0^+$ transition in ^{202}Pb is presented, along with that of transitions in other Pb products.

3.4.1 γ -ray Production Cross Section of the $2^+ \rightarrow 0^+$ Transition in ^{202}Pb

Sitting at $Z = 82$, ^{202}Pb is an ideal probe of the physics of even-even spherical nuclei. With extensive existing information in its structure, this work heavily relies on the evaluation of Kondev as seen in the Evaluated Nuclear Structure Data File (ENSDF) [47].

The ground state of ^{202}Pb has a half-life of $5.25 \times 10^4(28)$ years with no evidence of any γ -ray emission following electron capture decay. There has therefore never been any direct measurement of its production cross section. There however exists extensive data for the production of $^{202\text{m}}\text{Pb}$, a spin isomer ($J^\pi = 9^-$) at 2169.83 keV with a half-life of 3.54(2) h. The stacked-target experiment performed by the TREND collaboration adds to the set of available data. The stacked-target experiment data analyzed by Dr. Jonathan Morrell of BNL determined that the independent cross section of $^{nat}\text{Tl}(\text{p},\text{x})^{202\text{m}}\text{Pb}$ is 71.2 ± 2.6 mb at proton energy 30.33 ± 0.80 MeV and 51.4 ± 2.6 mb at proton energy 50.9 ± 1.6 MeV.

As will be discussed in the following sections, the measurement of the γ -ray production cross section of the yrast $2^+ \rightarrow 0^+$ transition in ^{202}Pb was simultaneously burdened and made possible by the existence of the isomeric state.

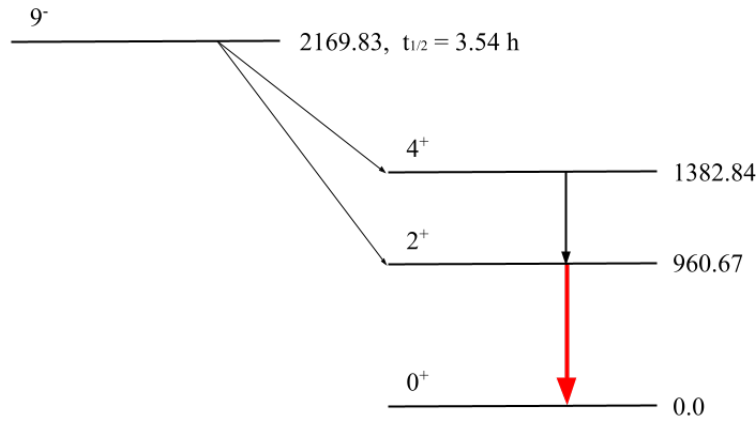


Figure 3.3: Low-lying levels in ^{202}Pb relevant to this work [47]. Transitions observed were drawn, with the $2^+ \rightarrow 0^+$ transition in bold and red.

The $2^+ \rightarrow 0^+$ Transition in ^{202}Pb

The population of the 0^+ ground state of ^{202}Pb can theoretically be achieved in the cascade of γ -ray transitions from various low-lying levels. However, as expected, most feeding into the ground state is from the 960.67 keV E2 transition initiated at the yrast 2^+ state of the nucleus. In fact, no other possible ground state feeding transitions were observed in this work - either likely due to significantly lower intensities than the $2^+ \rightarrow 0^+$ transition given the limits of detection efficiency, or the high possibility that the initial levels do not actually exist (see section 3.4.2 for details). The unobserved transitions are summarized in Table 3.1.

Table 3.1: **Unobserved** possible ground-state-feeding transitions in ^{202}Pb [47].

E_γ	E_i	J_i^π	Note
1382.8	1382.84	4^+	
1584	1584	2^+	Level likely does not exist
1657	1657	2^+	Level likely does not exist
1658.0	1658.0	0^+	
1798	1798	$(2)^+$	Level likely does not exist
1815	1815	No assignment	Level likely does not exist
1862	1862.0	0^+	
1915	1915.12	4^+	
2159	2159.0	0^+	

The measurement of the $2^+ \rightarrow 0^+$ transition is complicated by the fact that it is not only populated in the prompt γ -ray cascade in ^{202}Pb but also in the decay of $^{202\text{m}}\text{Pb}$, as shown in Figure 3.3, with intensity 90.1(4) %. The feeding of the transition via prompt reactions and radioactive decay must be deconvolved to measure the γ -ray production cross section of the 960.67 keV transition.

Deconvolving the Prompt Production and Decay Feeding

To remove the effect of angular dependency in γ -ray production, only spectra taken from the ORTEC GEM detector which sat at 90° at all times were used for the following process.

The prompt and decay contributions to the observed counts in the 960.67 keV peak can be separated by exploiting the properties of linearity in the differential equations governing activity build-up of radioisotopes.

The change in number of $^{202\text{m}}\text{Pb}$ nuclei during the irradiation is described by the equation of simultaneous production and decay,

$$\frac{dN_{\text{Pb-202m}}(t)}{dt} = R_{\text{Pb-202m}}(t) - \lambda_{\text{Pb-202m}}N_{\text{Pb-202m}}(t) \quad (3.3)$$

, where $N(t)$ is the number of nuclei present, λ is the decay constant, and $R(t)$ is the production rate. At $t = 0$, which is the start of each run, $N_{\text{Pb-202m}} = 0$. The production rate of $^{202\text{m}}\text{Pb}$ is given by

$$R_{\text{Pb-202m}}(t) = I(t) (\rho R) \sigma_{\text{isomer}} \quad (3.4)$$

, where $I(t)$ is the current of the incident proton beam, (ρR) is the areal density of the thallium foil, and σ_{isomer} is the production cross section of $^{202\text{m}}\text{Pb}$. With the relation that activity $A = \lambda N$, the number of counts in the 960.67 keV peak contributed by the decay of $^{202\text{m}}\text{Pb}$ during spectrum measurement time interval $[t_1, t_2]$ is

$$N_{\text{decay}} = \left(\int_{t_1}^{t_2} A_{\text{Pb-202m}}(t) \cdot \mathcal{I} dt \right) \times \varepsilon_{\text{detection}} \quad (3.5)$$

, where $\mathcal{I}$ is the intensity of the 960.67 keV γ -ray in the decay of $^{202\text{m}}\text{Pb}$ and $\varepsilon_{\text{detection}}$ is the detection efficiency.

The prompt contribution to the number of counts in the 960.67 keV peak during spectrum measurement time interval $[t_1, t_2]$ can be described simply as:

$$N_{\text{prompt}} = \left(\int_{t_1}^{t_2} I(t) \sigma_{\gamma} (\rho R) dt \right) \times \varepsilon_{\text{detection}} = Q(t_1, t_2) \sigma_{\gamma} (\rho R) \varepsilon_{\text{detection}} \quad (3.6)$$

, where $Q(t_1, t_2)$ is integrated proton charge in $[t_1, t_2]$, and σ_{γ} is the γ -ray production cross section of the 960.67 keV transition.

In every spectrum recorded, what is measured is the total number of counts in the 960.67 keV peak, *i.e.*,

$$N_{\text{total}} = N_{\text{decay}} + N_{\text{prompt}} \quad (3.7)$$

The current integrator provided us with a current reading every second during the irradiation. After correcting for the aforementioned data acquisition “efficiency” with ε_{DAQ} , we can fit all available spectra for each run to Equation 3.7, yielding the ratio of the yrast $2^+ \rightarrow 0^+$ partial γ -ray cross section relative to the $^{202\text{m}}\text{Pb}$ production cross section:

$$\begin{aligned} \frac{\sigma_{\gamma}}{\sigma_{\text{isomer}}} &= 2.99 \pm 0.48 \quad \text{for } E_p = 50 \text{ MeV} \\ &= 10.1 \pm 2.3 \quad \text{for } E_p = 30 \text{ MeV} \end{aligned} \quad (3.8)$$

However, the σ_{γ} shown in Equation 3.8 is not an angle-averaged production cross section for the 960.67 keV γ -ray, but a value specific to 90° ; it will hereafter be referred to as $\sigma_{\gamma}(90^\circ)$. Given the measured values of σ_{isomer} from the stacked-target experiment, it can be calculated that, for the $2^+ \rightarrow 0^+$ transition in ^{202}Pb ,

$$\begin{aligned} \sigma_{\gamma}(90^\circ) &= 154 \pm 26 \quad \text{mb for } E_p = 50 \text{ MeV} \\ &= 720 \pm 160 \quad \text{mb for } E_p = 30 \text{ MeV} \end{aligned} \quad (3.9)$$

Angular Dependence in Prompt γ -Ray Production

To obtain an angle-averaged production cross section for the prompt 960.67 keV transition, spectra collected using the moving Clover detector was utilized. Swung to angles 60° , 70° , 80° , and 90° during the course of the run, the angular dependence of γ -ray productions was captured.

However, since the 960.67 keV γ -ray is emitted both in the decay of $^{202\text{m}}\text{Pb}$ and the prompt cascade of γ transitions, the angular dependence of the prompt 960.67 keV transition could not be directly observed from the Clover spectra. To perform the aforementioned separation of decay and prompt contributions to the peak required long counting intervals at the same detector position, which was not the case for the Clover detector. To mitigate this, the angular dependence of the $2^+ \rightarrow 0^+$ transition in ^{200}Pb was used to approximate the behavior of the $2^+ \rightarrow 0^+$ transition in ^{202}Pb (*i.e.*, the 960.67 keV transition). There is no decay component to the $2^+ \rightarrow 0^+$ transition in ^{200}Pb .

This approximation is justified by the increasing isotropy of neutron emission in (p,xn) reactions as x increases. During irradiation, ^{202}Pb is produced via the $^{203}\text{Tl}(\text{p},2\text{n})$ reaction or the $^{205}\text{Tl}(\text{p},4\text{n})$ reaction; ^{200}Pb is produced via the $^{203}\text{Tl}(\text{p},4\text{n})$ reaction or the $^{205}\text{Tl}(\text{p},6\text{n})$ reaction. While the incident proton energies of this work of 30 and 50 MeV are within the range of inducing pre-equilibrium reactions, differences in the angular distribution of neutron emission are not expected to be particularly pronounced for the second, fourth, or sixth neutron emitted.

The γ -ray production cross section of the 1026.61 keV ($2^+ \rightarrow 0^+$) transition at 60° , 70° , and 80° were ratioed against that at 90° . Exploiting the symmetry of the system, the ratios were fitted to the first three even terms of the Legendre polynomial, *i.e.*,

$$\begin{aligned} P_n(\cos \theta, c_i) &= c_1 P_0(\cos \theta) + c_2 P_2(\cos \theta) + c_3 P_4(\cos \theta) \\ &= c_1 + \frac{c_2}{2}(3 \cos^2 \theta - 1) + \frac{c_3}{8}(35 \cos^4 \theta - 30 \cos^2 \theta + 3) \end{aligned} \quad (3.10)$$

, where θ is the angle of detection and c_i are the free parameters. An example of the fit is shown in Figure 3.4. As one can see, the angle of detection was quite large since the surface of the Clover detector had a width of 10 cm. To account for this, the fit was performed via orthogonal distance regression, and the uncertainties of the fitted parameters, Δc_i , were taken as one standard deviation to the fitted parameters.

For angle θ , the uncertainty to the fit is calculated as

$$\Delta P_n(\cos \theta, c_i) = |P_n(\cos \theta, c_i + \frac{\Delta c_i}{2}) - P_n(\cos \theta, c_i - \frac{\Delta c_i}{2})| \quad (3.11)$$

To obtain an angular-averaging factor for the transition, which would then be applied to $\sigma_\gamma(90^\circ)$ of the 960.67 keV transition, it is calculated as

$$\eta_\gamma = \frac{\int_0^\pi w(\theta) P_n(\cos \theta, c_i) \sin \theta d\theta}{\int_0^\pi w(\theta) \sin \theta d\theta} \quad (3.12)$$

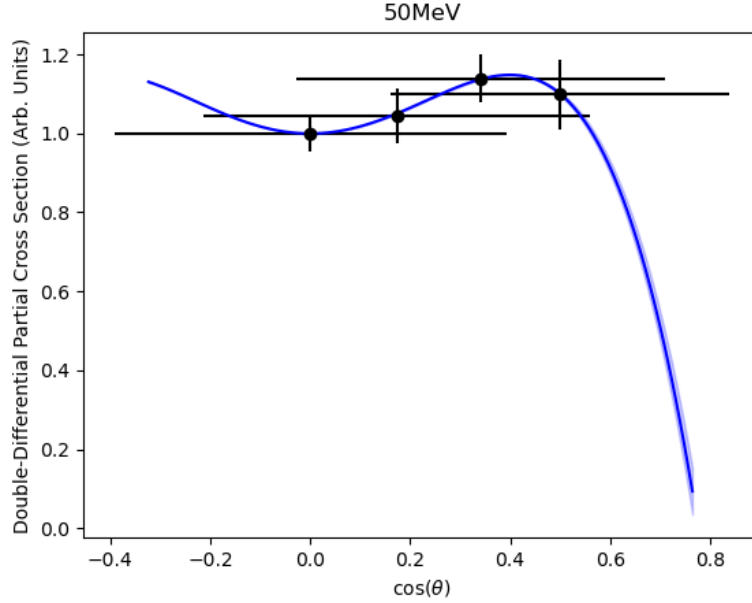


Figure 3.4: Fit to the angular distribution of the 1026.61 keV transition in ^{200}Pb at incident proton energy 50 MeV.

, where $w(\theta)$ is the weight to the average, defined by the relation

$$w(\theta) = 1/\Delta P_n(\cos \theta, c_i) \quad (3.13)$$

It was calculated that

$$\begin{aligned} \eta_\gamma &= 1.049 \pm 0.010 \quad \text{for } E_p = 50 \text{ MeV} \\ &= 0.913 \pm 0.012 \quad \text{for } E_p = 30 \text{ MeV} \end{aligned} \quad (3.14)$$

Finally, it is concluded that, the angular-averaged production cross section of the 960.67 keV transition,

$$\begin{aligned} \sigma_\gamma &= \sigma_\gamma(90^\circ) \times \eta_\gamma = 161 \pm 27 \quad \text{mb for } E_p = 50 \text{ MeV} \\ &= 660 \pm 150 \quad \text{mb for } E_p = 30 \text{ MeV} \end{aligned} \quad (3.15)$$

These angle-averaged cross sections are the appropriate ones to use in comparison to Talys calculations, since the code package does not include a treatment of the angular distribution of the emitted γ -rays caused by alignment of the magnetic substates in the nuclear reaction.

3.4.2 The Structure of ^{202}Pb

The current evaluation of the ^{202}Pb structure stems from a wide span of measurements including observations of decay from various parent nuclei and reactions with different combinations of incident particle and target material. The measurement called to attention here

is of the transfer reaction $^{204}Pb(p,t)$ by Lanford [48] with XREF flag J in the ENSDF evaluation [47]. In Lanford, the existence of the levels of energies 1584, 1657, 1798, and 1815 keV was suggested; however, as previously mentioned in Table 3.1, there is no indication of the existence of the levels in the measurements performed for this work, or any other work of prompt γ -spectroscopy. Given that a spin of 2^+ was assigned for three of the four levels (the 1815 keV level had no spin assignment), γ -ray production cross sections comparable to that of the 960.67 keV γ -ray are expected. None of these transitions, however, were seen. In Lanford, while measurements were performed for (p,t) reactions with $^{208,207,206,204}Pb$ targets, the author clearly stated that while the $^{208,207,206}Pb$ targets were all of higher than 97% enrichment, the ^{204}Pb target was only 90% enriched [48]. Misidentification of transitions in ^{202}Pb could have stemmed from the impurities in target material or contamination from environmentally abundant low-A nuclei such as C, N, or O.

The inclusion of spurious nuclear levels in ENSDF has been known for many years with Demidov *et al.* pointing out the non-existence of over 120 levels in 34 nuclei [49]. Notifying that these levels likely do not exist is not only important in improving the evaluation of the ^{202}Pb structure but also crucial in reaction modeling, discussed in Section 3.5.

3.4.3 Other γ -ray Production Cross Sections

In addition to the 960.67 keV transition in ^{202}Pb , quantification was achieved for multiple prompt γ -ray transitions in other lead isotopes of $198 \leq A \leq 203$. Transitions in even-A Pb nuclei were dominated by only a few yrast transitions. Figure 3.5 below is a sample of the gamma spectra measured in this work. Information of the transitions and measured cross sections are summarized in Table 3.2. The transitions presented here were those verified by the simultaneous observation of other transitions from or into shared levels, clearly resolved from the abundance of peaks and non-negligible noise in spectra, and whose strength is supported by theoretical calculations.

For each prompt γ -ray, using spectra collected with the ORTEC GEM detector, its production cross section at 90° can be calculated as

$$\sigma_\gamma(90^\circ) = \sigma_{960.67 \text{ keV}}(90^\circ) \times \frac{(N_c^\gamma / \varepsilon_{\text{detection}}(E_\gamma) / I_{\text{att}}(E_\gamma))}{(N_c^{960.67 \text{ keV}} / \varepsilon_{\text{detection}}(960.67 \text{ keV}) / I_{\text{att}}(960.67 \text{ keV}))} \quad (3.16)$$

, where $\sigma_{960.67 \text{ keV}}(90^\circ)$ is the production cross section of the 960.67 keV γ -ray of ^{202}Pb at 90° , $N_c^{960.67 \text{ keV}}$ is the number of counts from the **prompt** contribution in the 960.67 keV peak (derived in Section 3.4.1), $\varepsilon_{\text{detection}}$ is the detection efficiency at the corresponding γ -ray energy, and I_{att} is the attenuation factor of the γ -ray traveling through 0.8 cm of aluminum.

To arrive at the angular-averaged γ -ray production cross section, the angular-averaging factor η_γ defined as in Section 3.4.1 is calculated and applied for each identified γ -ray, with

$$\sigma_\gamma = \sigma_\gamma(90^\circ) \times \eta_\gamma \quad (3.17)$$

All values of $\sigma_\gamma(90^\circ)$ and σ_γ measured are presented in Table 3.2. Information on transition identification and initial and final levels are discussed below.

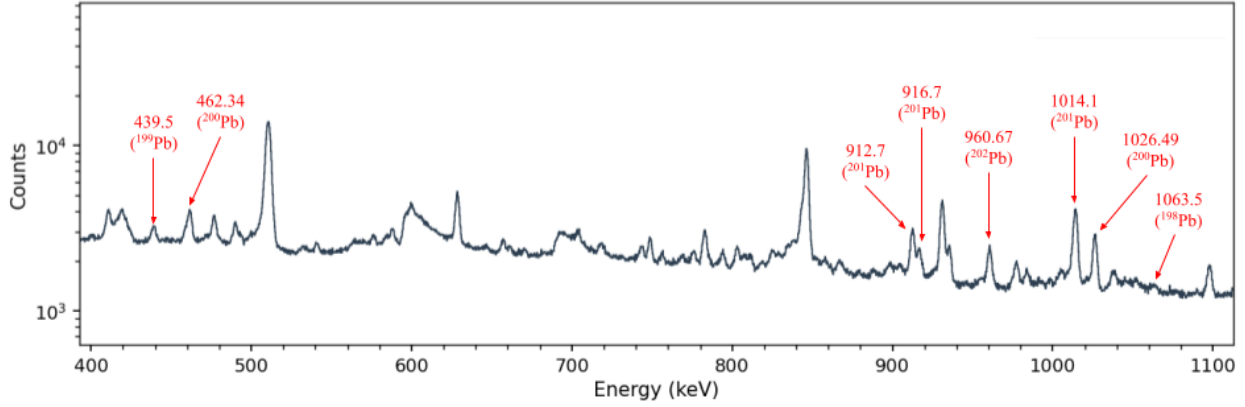


Figure 3.5: Sample of gamma spectra, from the ORTEC GEM detector with incident proton energy 50 MeV.

^{203}Pb [50]

At $E_p = 30$ MeV, the 869.9 keV E2 transition from the 896.9 keV level ($J^\pi = 9/2^-$) into ground state ($J^\pi = 5/2^-$) is observed.

^{202}Pb [47]

In addition to the 960.67 keV E2 transition from the yrast 2^+ state to the 0^+ ground state, which is the single most important transition in this work, the yrast $4^+ \rightarrow 2^+$ transition was also identified. However, at both $E_p = 50$ and 30 MeV, this 422.13 keV γ -ray was buried in a multiplet with at least 5 to 7 other γ -ray peaks, simply unable to be isolated. Its production was therefore not quantified.

^{201}Pb [51]

At both $E_p = 50$ and 30 MeV, three prompt transitions in ^{201}Pb were quantified. This includes the 1014.1 keV E2 transition from the 1014.2 keV level of $J^\pi = 9/2^-$. This is one of the strongest transition visible on the spectrum, with its existence corroborated by other measurements. The two other transitions measured are the 916.7 keV E2+M1 transition and the 912.7 keV E2 transition. Originating from the 1545.8 keV level of $J^\pi = 15/2^+$ and the 1541.8 keV level of $J^\pi = 17/2^+$ respectively, they share the final level of 629.1 keV with $J^\pi = 13/2^+$.

^{200}Pb [52]

At both $E_p = 50$ and 30 MeV, both the yrast $4^+ \rightarrow 2^+$ and $2^+ \rightarrow 0^+$ E2 transitions were identified. The $4^+ \rightarrow 2^+$ transition has γ -ray energy 462.34 keV, and the $2^+ \rightarrow 0^+$ transitions

has γ -ray energy 1026.61 keV. However, due to the lack of statistics, the $4^+ \rightarrow 2^+$ transition was not quantified for $E_p = 30$ MeV. The yrast 4^+ state has level energy 1488.3 keV, and the 2^+ state has level energy 1026.1 keV.

^{199}Pb [53]

The 439.5 keV E2 transition from the 1842.1+x keV level with $J^\pi = (21/2^+)$ to the 1402.5+x keV level with $J^\pi = (17/2^+)$ was clearly observed at $E_p = 50$ MeV. Given the parenthesis around the spin-parity assignments and the “+x” for level energies, it is obvious that there exists still a lot of uncertainties in the evaluation of the structure of ^{199}Pb . This transition was therefore excluded from the process of parameter adjustments for reaction modeling, discussed in the following section.

^{198}Pb [54]

At $E_p = 50$ MeV, the yrast $2^+ \rightarrow 0^+$ E2 transition from the 1063.5 keV level into the ground state level is quantified.

Table 3.2: γ -ray production cross sections, in units of mb, of observed transitions [50, 47, 51, 52, 53, 54].

Isotope	E_γ	$E_p = 50$ MeV		$E_p = 30$ MeV	
		$\sigma_\gamma(90^\circ)$	σ_γ	$\sigma_\gamma(90^\circ)$	σ_γ
^{203}Pb	896.9	-	-	249 ± 59	279 ± 66
^{202}Pb	960.67	154 ± 26	161 ± 27	720 ± 160	660 ± 150
^{201}Pb	916.7	86 ± 15	87 ± 15	98 ± 23	96 ± 22
^{201}Pb	912.7	194 ± 33	198 ± 34	203 ± 47	190 ± 44
^{201}Pb	1014.1	432 ± 73	458 ± 78	870 ± 200	990 ± 230
^{200}Pb	462.34	140 ± 29	151 ± 31	-	-
^{200}Pb	1026.49	209 ± 36	220 ± 37	25.7 ± 6.0	23.4 ± 5.5
^{199}Pb	439.5	84 ± 15	89 ± 15	-	-
^{198}Pb	1063.5	17.4 ± 3.2	17.8 ± 3.3	-	-

3.5 Charged-Particle Reaction Modeling with Talys-2.0

As mentioned, this work was complemented by stack-target measurements of $^{nat}\text{Tl}(\text{p},\text{x})$ performed at LBNL, LANL, and BNL, covering incident proton energies from reaction thresholds up to 200 MeV. A Talys parameter optimization process was applied using the cross section

measurements of said stacked-target measurements by Dr. Wilson Lin of BNL. It should be noted that the optimization was performed using neural networks in this case, similar to what is presented in Lin *et al.* [55]. The outcome of the parameter adjustments is included in Table 3.3.

Table 3.3: Optimized parameters relevant to this work. Parameters determined by Dr. Lin are presented in column “Lin”. Parameters ultimately adopted in this work are also presented (discussed in following sections). Default values to all parameters are 1.

Category	Adjusted Parameter	This Work	Lin
Level Density	ldmodel	2	2
	Rspincut	0.42	1.064765
Optical Model Potential	rvadjust n	1.	1.024657
	avadjust n	1.	1.170162
	rvadjust p	1.	0.974305
	avadjust p	1.	0.896286
	w1adjust n	1.	2.799852
	w2adjust n	1.	0.400392
	w1adjust p	1.	3.74766
	w2adjust p	1.	0.344649

In stacked-target experiments, while extensive residual production cross section measurements can be used in Talys parameter optimization, no information could be gathered for the production of long-lived or stable isotopes. The observed prompt γ -ray transitions in this work provided significant insight into the adjustments to discrete levels, angular momentum distribution in level densities, and ultimately a discussion of the convolution of optical model potentials and level densities in Hauser-Feshbach calculations.

3.5.1 Discrete Levels in Talys

Perhaps even more important than the global adjustments to level densities, the discrete level scheme should be as accurate as possible since it governs the γ -ray decay into lower-lying levels of residual nuclei. Where evaluated discrete level data, including energy and spin-parity assignments, are available in databases such as ENSDF, they are included in the editable discrete level files in Talys [37].

As previously discussed, multiple levels included in the ^{202}Pb structure evaluation in ENSDF were not observed in this work and hence should not be populated in the theoretical calculations of γ -ray cascades in the ^{202}Pb nucleus. This is achieved by increasing the spin of said levels to 21 in the discrete level file, minimizing their population relative to other nearby levels of similar spins that evidently exist.

The number of discrete levels in each residual nucleus incorporated was also increased from the default 10 to 25 via the keyword `maxlevelsres`, to invoke the explicit calculation of production cross sections of γ -rays observed in this work.

3.5.2 The Spin Cut-Off Parameter for Level Densities in Talys

As discussed in Chapter 2, both phenomenological and microscopic descriptions of level densities are available in Talys. When a phenomenological model is invoked, further adjustments to the angular momentum distribution of the level density are permitted. As shown in Table 3.3, `ldmodel 2`, the Back-shifted Fermi gas Model (BFM), was chosen in the set of optimized parameters. In BFM, spin J and parity π are accounted for as in Equation 3.18 [37]:

$$\rho(E_x, J, \pi) = \frac{1}{2} \frac{2J+1}{2\sigma^2} \exp \left[-\frac{(J+1/2)^2}{2\sigma^2} \right] \rho_{\text{tot}}(E_x) \quad (3.18)$$

, where ρ_{tot} is the total BFM level density, and σ^2 is the spin cut-off parameter.

Angular Momentum Distribution in Level Densities

The width of the angular momentum distribution in level densities is determined by the spin cut-off parameter, as shown in Equation 3.18. In Talys, the functional form of σ^2 incorporates effects from low-lying discrete levels up into the continuum and is defined as [37]:

$$\begin{aligned} \sigma^2(E_x) &= \sigma_d^2 && \text{for } 0 \leq E_x \leq E_d \\ &= \sigma_d^2 + \frac{E_x - E_d}{S_n - E_d} (\sigma_F^2(S_n) - \sigma_d^2) && \text{for } E_d \leq E_x \leq S_n \\ &= \sigma_F^2(E_x) && \text{for } E_x \geq S_n \end{aligned} \quad (3.19)$$

, where σ_F^2 is the level density of the continuum, σ_d^2 is the level density of the low-lying discrete levels, E_x is the excitation energy, S_n is the neutron separation energy, and E_d is the energy in the middle of the lower discrete level and the upper level.

The `Rspincut` parameter is a global multiplier to $\sigma^2(E_x)$, with default value 1 and a variable range of 0.1-10.

Adjustment to the Rspincut Parameter

All transitions listed in Table 3.2, excluding the 439.5 keV transition in ^{199}Pb , were used in the χ^2 -minimization process for `Rspincut` adjustments. The 439.5 keV transition was not used due to significant uncertainties in the evaluation of the structure of ^{199}Pb , from level energies to spin-parity assignments [53]. To make the optimization process in this work independent of the production cross section of $^{202\text{m}}\text{Pb}$ measured in the stacked-target experiment, the goodness-of-fit parameter depends solely on ratios of the γ -ray production cross sections to the $^{202\text{m}}\text{Pb}$ production cross section. For each prompt γ -ray transition,

$$s_i = \sigma_i^\gamma / \sigma_{\text{isomer}} \quad (3.20)$$

, where σ_i^γ is the γ -ray production cross section, and σ_{isomer} is the production cross section of $^{202\text{m}}\text{Pb}$.

The goodness-of-fit parameter χ^2 is defined as:

$$\chi^2 = \sum_{i=1}^{N_p} w_i \left(\frac{s_i^T - s_i^E}{\Delta s_i^E} \right)^2 \quad (3.21)$$

, where s_i^T is the Talys-calculated ratio, s_i^E is the experimentally-measured ratio, Δs_i^E is the uncertainty in the measured ratio, and w_i is the assigned weight. All transitions were assigned weight $w_i = 1$ except for the prompt 960 keV transition in ^{202}Pb , which was assigned weight $w_i = 5$. This is due to not only the utmost significance of this transition in the motivation of this work, but also the relative accuracy in measurement, since the quantification of all other γ -rays relied on that of this γ -ray.

Two cases were considered in the adjustment of **Rspincut** for χ^2 -minimization, where the default or values optimized by Lin for optical model potential (OMP) parameters were employed, as seen in Figure 3.6. As shown in Table 3.3, significant adjustments were made to parameters affecting the OMP, particularly for **w1adjust** and **w2adjust** for both neutrons and protons. In the case where the optimized parameters for OMP were applied, χ^2 was minimized at **Rspincut** = 2.3, signifying the widening of the angular momentum distribution by more than 2 times. In contrast, where the default values were used for all OMP parameters, the minimum in χ^2 is found when **Rspincut** is set to 0.42. The latter value is in agreement with what was concluded not only in the past TREND work of Apgar *et al.* [6], but also the publication of Rodrigo *et al.* [56], where more than 12,000 isomeric ratios from more than 900 light-charged-particle-induced reactions were investigated.

3.5.3 Level Density and Optical Model Potential in Hauser-Feshbach Calculations

Calculations of cross sections in Talys are governed first and foremost by the inputs to the optical model and the nuclear structure, including the level density, as shown in Figure 3.7. Harkening to discussions in Section 2.5.1 of the previous chapter, it was derived that the Hauser-Feshbach expression for energy-differential cross sections is given by:

$$\frac{d\sigma_{ab}^{HF}}{dE_b} = \pi\lambda \sum_{J\Pi} \omega_a^J \frac{\sum_{\{j_a\}} T_a}{\sum_c \sum_{\{n_c\}} \sum_{\{j_c\}} T_c} \sum_{I_b\Pi_b} \sum_{\{j_b\}} T_b \rho(E_b, I_b, \Pi_b) \quad (3.22)$$

, where T_i are the calculated transmission coefficients, $\{j_i\}$ represents the set of angular momenta, $\{n_i\}$ is the set of states, Π is the parity of the compound system, and $\rho(E_b, I_b, \Pi_b)$ is the level density of the final states.

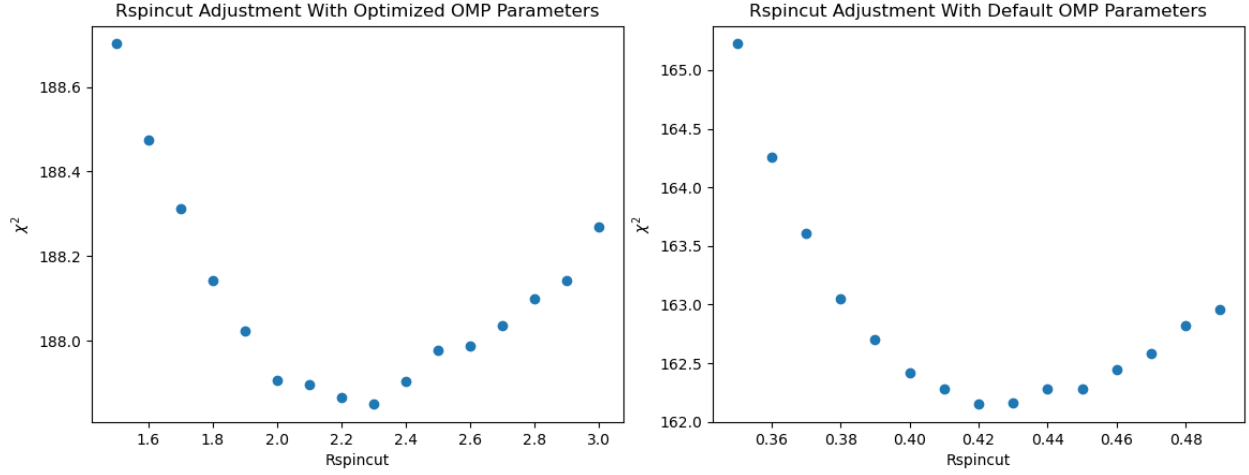


Figure 3.6: χ^2 of varying `Rspincut` values with optimized (left) and default (right) OMP parameters.

Simply from Equation 3.22, it is obvious that the changes made to the optical model potential and level density are not independent, as the transmission coefficients are derived invoking the optical model. As demonstrated, experimental outcomes of this work are best replicated by either significantly increasing the imaginary well depth of the optical potential (optimized OMP parameters of Lin) while doubling the spin cut-off parameter, or decreasing the width of angular momentum distributions in the level density by roughly half. The changes to the OMP parameters suggested by Lin's optimization are visualized in Figure 3.8.

It is difficult to physically justify the 2.3-times widening of the width angular momentum distribution of the quasi-continuum that had to be performed when the OMP parameters optimized by Lin were used. Given the agreement in the resulting best `Rspincut` ≈ 0.4 among Rodrigo *et al.* [56], Apgar *et al.* [6], and this work, it could be argued that the drastic adjustments to increase the probability of projectile absorption (significant increases to `w1adjust` from default) is compensating for the overestimation of the spin cut-off parameter.

3.6 Production Cross Section of ^{202g}Pb

To achieve the quantification of ^{202g}Pb production, the measured γ -ray production cross section of the $2^+ \rightarrow 0^+$ transition in ^{202}Pb and the Talys-calculated outcomes were incorporated.

While the ground state of ^{202}Pb is mostly populated by the yrast $2^+ \rightarrow 0^+$ transition, direct population from levels in the quasi-continuum is not negligible, as visualized in Figure 3.9. While transitions of such could not be individually measured, Talys provided theoretical

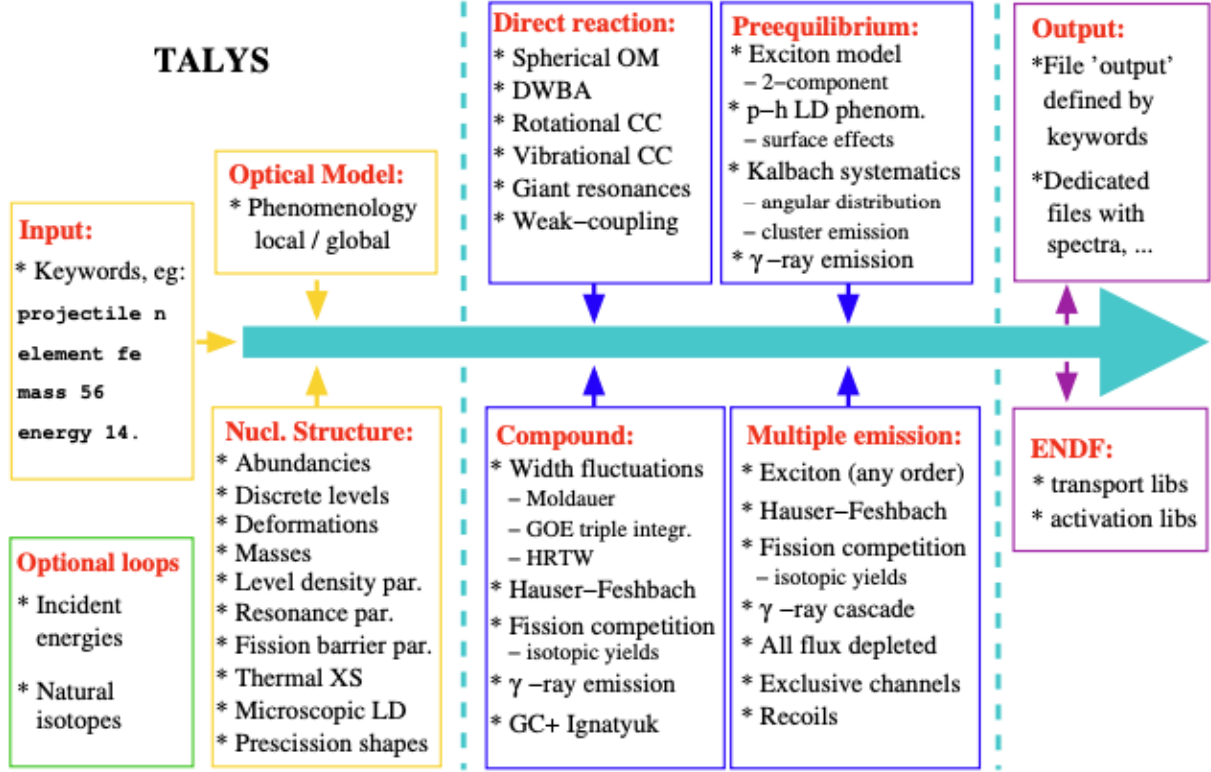


Figure 3.7: Models in Talys, taken from the Talys manual [37].

calculations of their total intensity compared to the yrast $2^+ \rightarrow 0^+$ transition. Adopting the R_{spincut} value of 0.42 and the default values for OMP parameters, Talys theorized that the ^{202}Pb ground state was populated through the yrast $2^+ \rightarrow 0^+$ transition approximately 88 % and 85 % of the time at 50 and 30 MeV incident proton energy, respectively. This is the ratio of the γ -ray production cross section of the $2^+ \rightarrow 0^+$ transition to the total production cross section of ^{202}gPb . Finally, by incorporating these values, the production cross section of ^{202}gPb is calculated as:

$$\begin{aligned} \sigma(^{202}\text{gPb}) &= \sigma_{960.67 \text{ keV}} \times \frac{\sigma^T(^{202}\text{gPb})}{\sigma_{960.67 \text{ keV}}^T} = 184 \pm 31 \quad \text{mb for } E_p = 50 \text{ MeV} \\ &= 770 \pm 180 \quad \text{mb for } E_p = 30 \text{ MeV} \end{aligned} \quad (3.23)$$

, where $\sigma(^{202}\text{gPb})$ is the measured γ -ray production cross section of the $2^+ \rightarrow 0^+$ transition as presented in Section 3.4, $\sigma^T(^{202}\text{gPb})$ is the Talys-calculated ^{202}gPb production cross section, and $\sigma_{960.67 \text{ keV}}^T$ is the Talys-calculated γ -ray production cross section of the $2^+ \rightarrow 0^+$ transition.

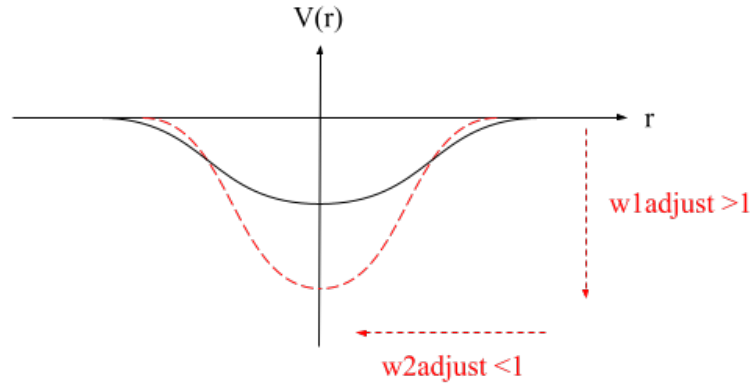


Figure 3.8: Visualization of parameter adjustments for the imaginary potential well.

3.7 Conclusions and Future Considerations

Motivated by the demand for the field of geochronology, an in-beam γ -ray spectroscopy measurement was performed at the 88-Inch Cyclotron of LBNL to characterize the production ^{202}gPb via the $^{nat}\text{Tl}(p,x)$ reaction pathway with incident proton energies of 30 and 50 MeV. Measurements of γ -ray production cross sections for transitions in $^{198, 199, 200, 201, 202, 203}\text{Pb}$ were made. No direct measurement has ever been made for the partial cross sections presented in this work.

The experimental measurements were further used to explore the effects of angular momentum distributions in level density models, through the optimization of the spin cut-off parameter in Talys-2.0. A deeper investigation into the simultaneous adjustments to the spin cut-off parameter and those of the optical model potential revealed that variance minimization approaches, a ML model in this case, will compensate for a truncated spin distribution through a dramatic increase in the depth of the imaginary optical model potential well. This result highlights the importance of direct measurements of reaction observables, such as the relative population of discrete states with differing spins.

Through Talys calculations with adjusted parameters, the ^{202}Pb ground-state feeding from the quasi-continuum was accounted for, yielding the total production cross sections of ^{202}gPb of 183 ± 31 mb and 770 ± 180 mb at 50 and 30 MeV incident proton energies, respectively.

Finally, with the current knowledge of problems in the data acquisition systems and challenges in data analysis, there are several improvements that can be made for future work of the same type. They include:

- Adding the cyclotron RF signal as a trigger for the data acquisition modules. This would provide a more accurate way to differentiate prompt and decay contributions to a transition populated by both mechanisms.

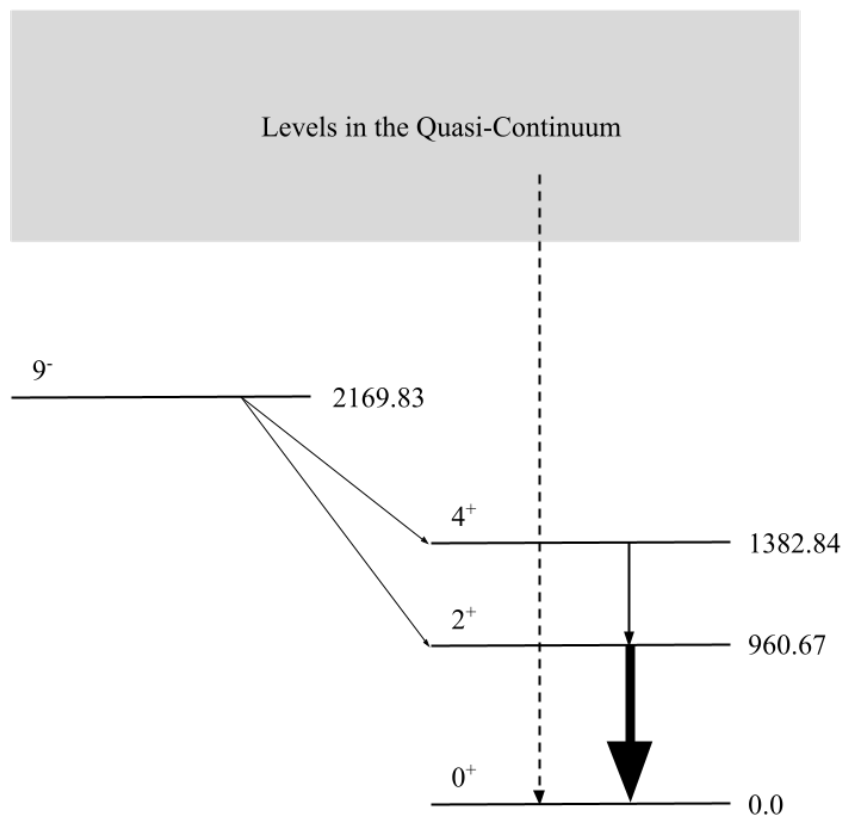


Figure 3.9: Rendering of Figure 3.3, with the notion of ground-state feeding from the quasi-continuum.

- Using targets that are of suitable thickness for in-beam γ -ray spectroscopy measurements. This would allow for the use of higher beam current, eliminating the need to assume current integrator behavior.
- Enhancing efficiency of the detection system to allow for coincidence γ -ray tracking. This would give more information into the structure of all produced nuclei, and guarantee the correct identification of all transitions.

Chapter 4

Conclusions

This dissertation encompasses the variability in experimental techniques, from established methods to innovative adaptations, to address the lack of high-energy charged particle reaction data for accelerator-based isotope production. These TREND campaigns provide the community with cross section data in need for the production of high-priority isotopes for various applications.

In high demand with evident impacts in brachytherapy, the field of isotope production is in need of experimental data for the assessment of high-energy production pathways of ^{103}Pd . Obtained via stacked-target irradiations at BNL and LANL, this dataset took another look at the excitation function of $^{\text{nat}}\text{Ag}(p,x)^{103}\text{Pd}$ below 100 MeV, and provided an extension of experimental data up to 200 MeV. Outcomes of the stacked-target irradiations for characterizing ^{103}Pd include cross section data for 24 $^{\text{nat}}\text{Ag}(p,x)$ reactions, 23 $^{\text{nat}}\text{Cu}(p,x)$ reactions, and 16 $^{\text{nat}}\text{Ni}(p,x)$ reactions, from proton energies 40 to 200 MeV. This measurement corroborated discoveries made in past TREND work that systematic uncertainties stemming from secondary particle induced reactions could be more pronounced than expected, calling for further scrutiny of uninformed employments of the stacked-target method [6].

To address the lack of production cross section data of ^{202g}Pb , a long-lived isotope used in the field of geochronology through mass spectroscopy, the TREND collaboration pivoted to γ -ray production cross section measurements of the $^{\text{nat}}\text{Tl}(p,x)$ reaction via in-beam γ -ray spectroscopy. With the assistance of theoretical calculations of Talys, this generated the first direct measurement of the ^{202g}Pb production cross section, with values 184 ± 31 mb and 770 ± 180 mb at proton energies 50 MeV and 30 MeV respectively. Additional γ -ray production cross sections measurements were made for transitions in lead isotopes of $198 \leq A \leq 203$.

Regardless of the motivations of the experiments, whether it be for cancer treatment or lunar formation dating, experimental cross section data are valuable gateways to understanding the fundamental frameworks of reaction physics. Theoretical calculations are important not only in providing the isotope production field with prediction capabilities for production campaigns, but also in the curation of evaluated nuclear data for all purposes related to nuclear physics. The TREND collaboration has been forwarding this aspect of the nuclear

data field via parameter optimizations with the tunable reaction modeling code Talys. Continuing the effort, cross section data generated in the stacked-target experiment of $^{\text{nat}}\text{Ag}(p,x)$ reactions were used to adjust parameters of level density, photon strength, pre-equilibrium reactions, and optical model potentials in Talys-2.0. Prompt γ -ray production cross section data from the in-beam measurements of $^{\text{nat}}\text{Tl}(p,x)$ reactions offered depth instead of span in parameter adjustments; the interplay of angular momentum distributions and optical model potentials was center to the discussion.

Two peer-reviewed publications are expected to originate from this dissertation, with the intention of incorporating all cross section data into EXFOR upon publication to guarantee data availability.

This dissertation merges the continuation of efficient and long-standing experimental efforts and the path towards higher accuracy in quantification of stable or long-lived products. While the entirety of this dissertation could be summed up into very specific applications in isotope production and niche discernment in nuclear reaction theory, I hope that it truly serves to nudge humankind towards the successes in curing illnesses and seeking ancient truths of our planet - all with the *magic of alchemy*.

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Appendix A

Stack Design and Foil Specifications

Specifications of each component of the stacks irradiated for the $^{\text{nat}}\text{Ag}(\text{p},\text{x})$ measurement are shown in Table A.1 and Table A.2.

Table A.1: Specifications the BNL stack. The proton beam travels through the stack in the order shown below, top to bottom. Uncertainties in measured areal density are given in least significant digits, that is, 97.08(2) means 97.08 ± 0.02 .

Foil ID	Compound	Thickness (μm)	Areal Density (mg/cm^2)
SS01 - Profile Monitor	SS	129	97.08(2)
Cu01	Cu	29	22.26(2)
Ag01	Ag	26	23.92(2)
Ni01	Ni	27	20.29(2)
Degrader 01	Cu	5219	4672
Cu02	Cu	29	22.09(2)
Ag02	Ag	27	24.99(2)
Ni02	Ni	27	21.59(2)
Degrader 02	Cu	4436	3969
Cu03	Cu	29	22.16(2)
Ag03	Ag	26	24.31(2)
Ni03	Ni	27	21.45(2)
Degrader 03	Cu	4428	3967
Cu04	Cu	29	22.15(2)
Ag04	Ag	30	29.23(2)
Ni04	Ni	27	21.91(2)
Degrader 04	Cu	4176	3743
Cu05	Cu	30	22.24(2)
Ag05	Ag	30	28.33(2)
Ni05	Ni	27	22.16(2)
Degrader 05	Cu	3723	3330

Continuation of Table A.1			
Cu06	Cu	29	22.15(2)
Ag06	Ag	27	25.75(2)
Ni06	Ni	28	22.48(2)
Degrader 06	Cu	3676	3285
Cu07	Cu	29	22.10(2)
Ag07	Ag	26	24.41(2)
Ni07	Ni	28	22.58(2)
SS02 - Profile Monitor	SS	128	98.19(3)

Table A.2: Specifications of the LANL stack, presented as in Table A.1.

Foil ID	Compound	Thickness (μm)	Areal Density (mg/cm^2)
SS01 - Profile Monitor	SS	53	39.99(1)
Cu01	Cu	31	22.02(2)
Ag01	Ag	26	25.09(2)
Ni01	Ni	29	22.64(2)
Degrader 01	Cu	814	676.3(2)
Cu02	Cu	30	22.01(2)
Ag02	Ag	26	25.94(2)
Ni02	Ni	28	22.17(2)
Degrader 02	Cu	545	432.3(1)
Cu03	Cu	30	22.04(2)
Ag03	Ag	26	25.29(2)
Ni03	Ni	29	22.38(2)
Degrader 03	Cu	814	680.8(1)
Cu04	Cu	30	22.21(2)
Ag04	Ag	26	25.66(2)
Ni04	Ni	29	21.50(2)
Degrader 04	Cu	524	434.5(1)
Cu05	Cu	30	22.09(2)
Ag05	Ag	27	26.30(2)
Ni05	Ni	29	20.32(2)
Degrader 05	Cu	830	680.4(2)
Cu06	Cu	30	22.02(2)
Ag06	Ag	23	23.07(2)
Ni06	Ni	27	21.62(2)
Degrader 06	Cu	529	436.6(1)
Cu07	Cu	30	22.13(2)
Ag07	Ag	23	23.21(2)

Continuation of Table A.2			
Ni07	Ni	29	21.88(2)
Degrader 07	Cu	1049	868.6(2)
Cu08	Cu	30	22.18(2)
Ag08	Ag	23	23.15(2)
Ni08	Ni	29	20.20(2)
Degrader 08	Cu	1042	870.1(2)
Cu09	Cu	30	22.02(2)
Ag09	Ag	23	23.18(2)
Ni09	Ni	28	22.58(2)
Degrader 09	Cu	1035	874.6(2)
Cu10	Cu	30	22.21(2)
Ag10	Ag	24	23.19(2)
Ni10	Ni	27	20.25(2)
SS02 - Profile Monitor	SS	54	40.09(1)

Appendix B

Decay Data Used for All Products of Stacked-Foil Experiment

Decay data used for silver products and monitor foil products are listed in Tables B.1 and B.2 respectively.

Table B.1: Decay data of Ag products used in this work.

Isotope	Half-life	Energy (keV)	I_γ (%)
$^{107}\text{Cd}[57]$	6.50(2) h	93.124	4.7(3)
$^{106\text{m}}\text{Ag}[58]$	8.28(2) d	221.701	6.6(3)
		228.633	2.10(10)
		374.46	0.26(4)
		406.182	13.4(4)
		418.55	0.33(6)
		429.646	13.2(4)
		450.976	23.2(7)
		601.17	1.61(9)
		616.17	21.6(6)
		679.64	0.64(4)
		680.42	1.54(8)
		703.11	4.47(18)
		717.34	28.9(8)
		748.36	20.6(6)
		793.17	5.9(3)
		804.28	12.4(5)
		808.36	4.0(4)
		824.69	15.3(4)

Continuation of Table B.1			
		847.03	2.8(6)
		847.27	1.6(5)
		874.81	0.33(4)
		956.22	0.47(8)
		1019.72	1.04(16)
		1045.83	29.6(10)
		1053.77	0.96(14)
		1121.59	0.57(6)
		1128.02	11.8(5)
		1199.39	11.2(5)
		1222.88	7.0(4)
		1394.35	0.12(4)
		1527.65	16.3(13)
		1565.4	0.48(4)
		1572.35	6.6(5)
		1722.76	1.40(18)
		1839.05	2.0(3)
^{105}Ag [59]	41.29(7) d	155.39	0.44(3)
		182.85	0.35(3)
		280.44	30.2(17)
		344.52	41.40(207)
		360.66	0.50(3)
		401.65	0.174(13)
		414.66	0.29(3)
		437.12	0.25(3)
		442.25	4.720(236)
		443.37	10.5(5)
		560.72	0.58(4)
		617.85	1.16(6)
		673.21	1.05(6)
		743.31	0.57(4)
		807.46	1.16(7)
		1087.94	3.85(17)
^{104}Cd [60]	57.7(10) m	559	6.3(8)
		625.7	2.2(3)
		709.3	19.5(24)
^{104}Ag [60]	69.2(10) m	740.5	7.2(9)
		857.9	10.4(14)

Continuation of Table B.1			
		863	6.9(9)
		908	4.4(6)
		923.3	6.9(9)
		925.9	12.5(15)
		941.6	25.0(23)
		1265.2	4.3(7)
		1526.6	7.1(9)
		1625.8	5.1(7)
$^{103}\text{Ag}[61]$	65.7(7) m	118.74	31.2(20)
		148.2	23.3(17)
		243.96	8.5(4) ¹
		266.86	13.3(9)
		1273.83	9.4(6)
$^{103}\text{Pd}[61]$	16.991(19) d	39.748	0.0683(34) ¹
$^{103}\text{Ru}[61]$	39.247(3) d	39.760	0.0692(11)
		497.085	91.0(12)
$^{102}\text{Rh}[62]$	207.3(3) d	475.06	46(4)
$^{101}\text{Pd}[63]$	8.47(6) h	269.67	33.5(6)
		296.29	100(0)
		320.74	2.94(15)
		453.7	3.15(12)
		565.98	17.9(4)
		590.44	62.8(10)
		723.75	10.2(6)
		992.84	4.9(3)
		1177.63	1.84(10)
		1202.04	7.9(3)
		1289.05	11.9(3)
$^{101\text{m}}\text{Rh}[63]$	4.34(1) d	306.857	81(4)
		545.117	4.3(3)
$^{100}\text{Pd}[64]$	3.63(9) d	42.08	7.0(6)
		74.78	48(4)
		84.00	52(4)
		126.15	7.8(7)

Continuation of Table B.1			
		158.87	1.66(14)
^{100}Rh [64]	20.5(3) h	446.153	11.98(11)
		588.343	5.01(4)
		590.792	1.089(13)
		822.654	21.09(17)
		1034.51	1.546(12)
		1107.223	13.57(11)
		1341.515	5.29(4)
		1362.152	15.39(12)
		1553.348	20.67(17)
		1559.554	0.999(9)
		1627.34	1.635(15)
		1929.811	11.61(12)
$^{99\text{m}}\text{Rh}$ [65]	46.2(16) m	89.6	1.75(18)
		250.9	0.52(5)
		276.6	1.67(13)
		322.1	0.90(12)
		340.8	72.0(21)
		486.1	0.66(8)
		617.8	12.3(11)
		685.6	0.86(14)
		719.3	1.25(7)
		734.1	0.45(5)
		920.0	0.77(6)
		936.6	2.25(11)
		1261.2	11.4(6) ¹
^{97}Rh [66]	30.7(6) m	421.55	74.63(3.73)
^{97}Ru [66]	2.83(23) d	215.7	85.62(428) ¹
		324.49	10.79(17)
^{96}Tc [67]	4.28(7) d	812.54	82(3)
		1126.85	15.2(12)
^{95}Ru [68]	1.643(13) h	336.40	69.9(5)
		626.83	17.8(5)
		1096.80	20.9(10)

Continuation of Table B.1			
^{95}Tc [68]	20.0(1) h	765.789	93.8(3)
		947.67	1.951(19)
		1073.71	3.74(4)
^{94}Ru [69]	51.8(6) m	367.2	75(8)
		891.2	25(5)
$^{94\text{m}}\text{Tc}$ [69]	52.0(10) m	1868.68	5.7(3)
		2740.1	3.5(3)
$^{93\text{m}}\text{Mo}$ [70]	6.86(7) h	263.049	57.4(11)
		684.693	99.9(8)
^{90}Mo [71]	5.56(9) h	122.37	64(3)
		257.34	77(4)
^{90}Nb [71]	14.60(5) h	141.178	66.8(7)
		1129.224	92.7(5)
		2186.242	17.96(17)
		2318.959	82.0(3)

Table B.2: Decay data of monitor foil products used in this work.

Isotope	Half-life	Energy (keV)	I_γ (%)
^{65}Zn [72]	243.93 (9) d	1115.539	50.12(7)
^{64}Cu [73]	12.7006(20) h	1345.77	0.472(5)
^{63}Zn [74]	38.49(5) m	669.62	8.3(5)
		962.06	6.53(32)
^{62}Zn [75]	9.197(20) h	548.35	15.3(14)
		596.56	26.0(13) ¹
^{61}Cu [76]	3.336(10) h	282.956	12.7(20)
		373.050	2.14(33)
		529.169	0.37(6)

¹Where no uncertainty is available from evaluation a relative 5% uncertainty is assigned.

Continuation of Table B.2			
		588.605	1.17(18)
		656.008	10.4(16)
		816.692	0.35(5)
		841.211	0.24(4)
		908.631	1.09(17)
		1099.56	0.27(4)
		1132.351	0.091(14)
		1185.234	3.6(6)
$^{60}\text{Cu}[77]$	23.7(4) m	826.4	21.7(11)
		1791.6	45.4(23)
$^{60}\text{Co}[77]$	1925.8(14) d	1173.228	99.85 (3)
$^{59}\text{Fe}[78]$	44.490(9) d	1099.245	56.5(9)
		1291.59	43.2(9)
$^{58}\text{Co}[79]$	70.86(6) d	810.7593	99.450(4972) ¹
		863.951	0.686(10)
		1674.725	0.517(10)
$^{57}\text{Ni}[80]$	35.60(6) h	127.164	16.7(5)
		1377.63	81.7(24)
		1757.55	5.75(20)
		1919.52	12.3(4)
$^{57}\text{Co}[80]$	271.74(6) d	122.06065	85.60(17)
		136.47356	10.68(8)
$^{56}\text{Ni}[81]$	6.075(10) d	158.38	98.8(10)
		269.5	36.5(8)
		749.95	49.5(12)
		811.85	86.0(9)
$^{56}\text{Co}[81]$	77.236(26) d	1037.843	14.05(4)
		1175.101	2.252(6)
		1238.288	66.46(12)
		1360.212	4.283(12)
		1771.357	15.41(6)
		2015.215	3.016(12)
		2034.791	7.77(3)

Continuation of Table B.2			
^{56}Mn [81]	2.5789(1) h	1810.726 2113.092	26.9(4) 14.2(3)
^{55}Co [82]	17.53(3) h	477.2 803.7 931.1 1316.6 1370 1408.5	20.2(17) 1.87(15) 75(4) ¹ 7.1(3) 2.9(3) 16.9(8)
^{54}Mn [83]	312.20(20) d	834.848	99.9760(10)
^{52}Mn [84]	5.591(3) d	744.233 1246.278	90.0(12) 3.32(4)
^{51}Cr [85]	27.704(3) d	320.0824	9.910(10)
^{49}Cr [86]	42.3(1) m	62.289 90.639 152.928	16.4(6) 53.2(19) 30.3(11)
^{48}Cr [87]	21.56(3) h	112.31 308.24	96.0(20) 100.0(20)
^{48}V [87]	15.974(3) d	944.13 2240.396	7.870(7) 2.333(13)
^{48}Sc [87]	43.71(9) h	1037.522	97.5(20)
^{46}Sc [88]	83.79(4) d	889.277 1120.545	99.9840(10) 99.9870(10)
^{45}Ti [89]	184.8(5) m	719.6	0.154(12)
$^{44\text{m}}\text{Sc}$ [90]	58.61(10) h	271.241	86.7(3)
^{43}Sc [91]	3.891(12) h	372.9	22.5(11)

¹Where no uncertainty is available from evaluation a relative 5% uncertainty is assigned.

Appendix C

Measured Cross Sections from Stacked-Foil Experiment

All measured production cross sections for Ag foil products are shown in Table C.1. All measured production cross sections for monitor foil products are tabulated in Tables C.2 and C.3.

Table C.1: $^{\text{nat}}\text{Ag}(\text{p},\text{x})$ cross sections measured in this work.

Production cross sections (mb)						
E_p (MeV)	187.1(21)	172.1(22)	158.6(23)	144.3(25)	129.9(27)	116.0(29)
	101.0(32)	91.2(11)	86.9(11)	83.9(11)	79.3(12)	76.1(12)
	71.2(13)	67.7(13)	60.8(14)	53.3(16)	44.8(18)	-
$^{107}\text{Cd}_{(i)}$	15.2(13)	14.1(11)	15.0(20)	19.4(31)	19.3(17)	23.3(37)
	25.7(19)	34.3(35)	29.0(25)	31.5(28)	34.9(29)	36.9(31)
	40.7(33)	43.1(34)	36.1(49)	62.6(50)	86(12)	-
$^{106\text{m}}\text{Ag}_{(i)}$	29.1(23)	34.2(27)	36.7(28)	39.3(29)	42.2(30)	46.0(32)
	52.9(36)	47.9(39)	48.5(38)	52.3(41)	54.6(42)	57.5(44)
	60.2(45)	63.0(46)	73.0(52)	93.5(65)	86.7(59)	-
$^{105}\text{Ag}_{(c)}$	91.0(73)	104.6(82)	111.6(85)	120.7(89)	131.8(95)	145(10)
	168(12)	162(13)	165(13)	179(14)	191(15)	203(15)
	227(17)	250(18)	296(21)	285(20)	224(15)	-
$^{104}\text{Cd}_{(i)}$	-	-	-	-	-	-
	-	-	30.0(47)	33.5(48)	36.2(48)	-
	42.6(51)	43.5(43)	-	55.9(42)	71.0(55)	-
$^{104}\text{Ag}_{(c)}$	51.6(43)	56.9(45)	63.0(52)	66.6(51)	71.6(54)	80.6(59)
	91.8(67)	-	111.3(98)	123(11)	137(11)	-
	145(12)	140(11)	-	187(14)	219(16)	-
$^{103}\text{Ag}_{(c)}$	48.8(42)	-	-	63.3(53)	67.2(52)	80.1(63)

Continuation of Table C.1						
	94.1(73)	101.4(89)	98.9(85)	100.9(86)	98.6(86)	97.5(85)
	106.8(87)	120(11)	131.3(97)	71.7(54)	3.67(37)	-
$^{103}\text{Pd}_{(c)}$	140(12)	149(14)	148(23)	178(18)	169(12)	183(19)
	217(32)	258(38)	240(24)	216(18)	214(23)	239(36)
	303(27)	263(26)	309(33)	155(12)	-	-
$^{103}\text{Ru}_{(c)}$	0.357(29)	0.347(28)	0.331(27)	0.279(21)	0.242(18)	0.192(15)
	0.151(12)	0.133(11)	0.105(11)	0.093(10)	0.1109(93)	0.0864(93)
	0.0809(78)	0.0555(97)	0.0358(90)	0.0219(46)	-	-
$^{102}\text{Rh}_{(i)}$	10.30(97)	11.03(99)	9.85(75)	8.76(70)	8.93(66)	8.18(93)
	7.36(70)	10.3(18)	4.1(12)	-	8.7(12)	-
	-	14.2(38)	-	-	-	-
$^{101}\text{Pd}_{(c)}$	53.9(44)	56.6(44)	59.3(55)	58.1(46)	59.6(44)	59.9(48)
	53.9(37)	47.6(39)	43.0(34)	39.3(31)	31.2(26)	26.2(22)
	19.7(15)	16.8(14)	17.3(13)	28.4(21)	19.1(14)	-
$^{101\text{m}}\text{Rh}_{(i)}$	30.2(28)	31.1(27)	37.2(52)	35.1(33)	33.1(33)	32.2(37)
	28.0(21)	34.0(30)	27.9(23)	27.0(23)	20.5(20)	19.5(20)
	13.9(13)	12.2(12)	10.83(97)	15.9(19)	7.92(92)	-
$^{100}\text{Pd}_{(c)}$	32.3(26)	34.1(27)	34.5(26)	34.4(25)	32.6(24)	31.3(22)
	20.5(14)	17.1(14)	12.7(10)	13.9(11)	12.19(94)	13.5(10)
	16.1(12)	17.6(13)	17.1(12)	5.28(39)	0.465(52)	-
$^{100}\text{Rh}_{(m+)}$	34.6(28)	35.3(28)	34.9(28)	34.8(26)	31.4(23)	29.5(22)
	26.1(18)	19.8(16)	19.6(16)	15.7(13)	15.2(12)	15.9(13)
	18.1(14)	18.3(15)	10.73(78)	1.32(14)	-	-
$^{99\text{m}}\text{Rh}_{(c)}$	-	27.6(39)	-	-	-	21.6(42)
	-	22.3(20)	22.0(19)	-	22.2(23)	18.8(19)
	10.9(12)	6.12(61)	0.332(24)	-	-	-
$^{97}\text{Rh}_{(c)}$	10.4(11)	9.60(91)	8.4(10)	7.09(79)	6.62(62)	5.43(52)
	1.29(27)	-	-	-	-	-
	-	-	-	-	-	-
$^{97}\text{Ru}_{(c)}$	27.0(23)	24.7(20)	24.3(21)	20.7(16)	15.8(12)	13.3(10)
	5.20(44)	4.02(33)	2.67(21)	2.41(19)	2.30(18)	2.55(20)
	2.96(23)	2.89(21)	1.300(96)	-	-	-
$^{96}\text{Tc}_{(m+)}$	8.52(68)	7.75(60)	6.40(51)	5.42(40)	4.12(30)	2.64(19)
	1.69(12)	1.46(12)	1.37(11)	1.205(95)	0.731(57)	0.396(33)
	-	-	-	-	-	-
$^{95}\text{Ru}_{(c)}$	9.95(93)	8.35(69)	7.6(11)	5.61(79)	3.00(55)	-
	-	-	-	-	-	-
	-	-	-	-	-	-
$^{95}\text{Tc}_{(c)}$	11.0(11)	10.18(84)	7.9(16)	5.49(86)	3.71(65)	-
	-	-	-	-	-	-
	-	-	-	-	-	-

Continuation of Table C.1						
$^{94}\text{Ru}_{(c)}$	-	2.34(43)	1.58(28)	-	-	-
	-	-	-	-	-	-
	-	-	-	-	-	-
$^{94\text{m}}\text{Tc}_{(c)}$	-	-	0.00136(24)	-	-	-
	-	-	-	-	-	-
	-	-	-	-	-	-
$^{93\text{m}}\text{Mo}_{(c)}$	1.94(16)	1.54(12)	1.17(11)	0.813(78)	0.530(93)	-
	-	-	-	-	-	-
	-	-	-	-	-	-
$^{90}\text{Mo}_{(c)}$	0.642(65)	0.323(32)	-	-	-	-
	-	-	-	-	-	-
	-	-	-	-	-	-
$^{90}\text{Nb}_{(c)}$	2.32(23)	1.63(13)	-	-	-	-
	-	-	-	-	-	-
	-	-	-	-	-	-

Table C.2: $^{\text{nat}}\text{Cu}(\text{p},\text{x})$ cross sections measured in this work.

Production cross sections (mb)						
E_p (MeV)	187.2(21)	172.2(22)	158.7(23)	144.4(25)	129.9(27)	116.0(29)
	101.1(32)	91.4(11)	87.1(11)	84.1(11)	79.5(12)	76.3(12)
	71.3(13)	67.8(13)	61.0(14)	53.5(16)	45.0(18)	-
$^{65}\text{Zn}_{(i)}$	1.37(14)	1.76(14)	1.74(15)	1.85(14)	2.29(18)	2.47(19)
	2.70(19)	2.49(20)	2.78(23)	2.69(22)	2.97(24)	3.17(26)
	3.51(27)	3.68(28)	3.98(30)	4.87(35)	5.70(40)	-
$^{64}\text{Cu}_{(i)}$	32.4(38)	33.4(45)	39.4(36)	40.0(47)	42.3(32)	45.3(32)
	47.1(35)	35.3(33)	39.0(32)	40.9(34)	40.9(32)	38.9(32)
	45.2(35)	46.4(37)	48.6(35)	51.0(46)	56.2(41)	-
$^{63}\text{Zn}_{(i)}$	4.72(38)	5.21(41)	5.12(55)	5.87(46)	6.01(49)	7.07(52)
	8.51(75)	7.4(13)	7.88(87)	8.6(11)	9.72(99)	10.2(13)
	11.1(11)	11.34(93)	13.9(15)	18.1(17)	27.2(21)	-
$^{62}\text{Zn}_{(i)}$	2.12(22)	2.43(25)	2.69(28)	2.89(28)	2.96(34)	3.47(36)
	4.04(44)	4.34(36)	4.69(38)	4.87(39)	5.26(41)	5.60(43)
	6.14(46)	6.63(50)	8.26(62)	10.39(75)	11.33(80)	-
$^{61}\text{Cu}_{(c)}$	33.0(29)	36.6(31)	39.1(32)	41.2(33)	44.8(35)	49.9(37)
	56.3(41)	51.7(43)	56.2(46)	57.7(47)	62.1(50)	63.4(51)
	71.4(56)	74.6(58)	79.8(60)	89.4(74)	120.4(88)	-
$^{60}\text{Cu}_{(c)}$	7.81(98)	9.87(88)	10.23(92)	10.24(86)	11.7(10)	12.9(10)
	15.1(11)	15.7(33)	20.0(32)	-	17.8(27)	17.3(14)

Continuation of Table C.2						
$^{60}\text{Co}_{(c)}$	19.3(18)	19.4(20)	25.9(26)	29.6(24)	13.1(10)	-
	12.5(12)	15.1(26)	13.9(15)	14.3(13)	14.2(18)	-
	14.7(18)	12.6(12)	14.8(15)	14.5(15)	13.5(12)	12.9(13)
$^{59}\text{Fe}_{(c)}$	14.6(18)	12.6(15)	13.4(10)	11.29(85)	8.52(63)	-
	1.40(11)	1.42(11)	1.36(10)	1.280(96)	1.238(93)	1.172(84)
	1.008(74)	0.830(69)	0.864(70)	0.852(68)	0.831(65)	0.852(66)
$^{58}\text{Co}_{(m+)}$	0.765(61)	0.743(56)	0.619(46)	0.350(27)	-	-
	42.7(42)	46.7(42)	48.5(43)	47.4(44)	50.2(40)	51.0(42)
	52.6(42)	46.7(44)	47.8(44)	47.2(44)	45.6(45)	44.1(41)
$^{57}\text{Ni}_{(c)}$	39.1(35)	36.3(31)	30.8(25)	32.7(24)	50.5(37)	-
	1.95(17)	2.03(17)	2.05(17)	2.10(17)	1.96(16)	1.87(15)
	1.62(12)	1.42(12)	1.30(11)	1.20(10)	1.078(92)	1.109(93)
$^{57}\text{Co}_{(i)}$	1.24(10)	1.41(12)	1.99(15)	1.95(15)	0.473(35)	-
	43.1(35)	45.0(36)	44.44(35)	44.9(33)	44.4(33)	45.7(33)
	42.3(29)	38.6(31)	38.5(31)	36.3(29)	35.1(27)	34.6(26)
$^{56}\text{Ni}_{(c)}$	34.9(26)	37.1(27)	46.6(33)	53.7(38)	24.1(16)	-
	0.185(24)	0.174(20)	0.162(20)	0.128(12)	0.125(13)	0.098(14)
	-	0.0803(77)	0.0747(67)	0.0832(71)	0.0897(82)	0.0994(87)
$^{56}\text{Co}_{(i)}$	0.0942(80)	0.0807(70)	0.0394(45)	-	-	-
	13.4(11)	13.8(11)	13.5(11)	13.7(10)	13.24(97)	12.43(90)
	11.26(78)	10.60(86)	11.51(92)	11.82(93)	13.1(10)	14.1(11)
$^{56}\text{Mn}_{(c)}$	13.9(10)	12.33(91)	5.89(43)	-	-	-
	2.26(28)	2.20(29)	2.17(25)	1.89(18)	1.73(21)	1.39(21)
	-	0.92(12)	0.92(11)	-	-	0.883(77)
$^{55}\text{Co}_{(c)}$	-	0.494(58)	-	-	-	-
	2.09(18)	2.10(18)	2.07(17)	1.94(17)	1.79(26)	1.64(20)
	1.70(24)	1.51(13)	1.45(12)	1.24(10)	0.882(88)	0.624(79)
$^{54}\text{Mn}_{(i)}$	0.286(27)	-	-	-	-	-
	17.9(14)	17.5(14)	16.1(12)	14.7(11)	13.18(95)	11.82(83)
	7.72(53)	6.29(51)	5.13(41)	4.36(34)	3.76(29)	3.69(29)
$^{52}\text{Mn}_{(c)}$	3.85(29)	4.29(32)	4.71(34)	2.83(21)	0.331(25)	-
	5.41(44)	4.85(38)	4.30(33)	3.62(42)	2.72(44)	1.97(25)
	1.79(30)	1.86(15)	1.33(11)	0.989(82)	0.510(43)	0.270(23)
$^{51}\text{Cr}_{(c)}$	0.0707(84)	0.0296(46)	-	-	-	-
	13.0(11)	10.71(86)	9.04(71)	7.35(56)	5.76(42)	4.73(36)
	1.92(14)	-	-	-	-	-
$^{48}\text{V}_{(c)}$	-	-	-	-	-	-
	2.48(22)	1.64(14)	1.22(12)	0.760(63)	-	-
	-	-	-	-	-	-
$^{48}\text{Sc}_{(c)}$	-	-	-	-	-	-
	-	0.109(21)	-	-	-	-

Continuation of Table C.2						
	-	-	-	-	-	-
	-	-	-	-	-	-
$^{46}\text{Sc}_{(m+)}$	0.607(49)	0.363(31)	0.244(20)	0.166(17)	-	-
	-	-	-	-	-	-
	-	-	-	-	-	-
$^{45}\text{Ti}_{(c)}$	97(16)	95(18)	-	90(17)	109(16)	-
	-	-	-	-	-	-
	-	-	-	-	-	-
$^{44\text{m}}\text{Sc}_{(c)}$	0.351(30)	0.145(17)	0.097(18)	-	-	-
	-	-	-	-	-	-
	-	-	-	-	-	-

Table C.3: $^{\text{nat}}\text{Ni}(\text{p},\text{x})$ cross sections measured in this work.

Production cross sections (mb)							
E_p (MeV)	187.1(21)	172.0(22)	158.5(23)	144.2(25)	129.8(27)	115.8(29)	100.9(32)
	91.1(11)	86.8(11)	83.8(11)	79.2(12)	76.0(12)	71.0(13)	67.5(13)
	60.6(15)	53.1(16)	44.6(18)	-	-	-	-
$^{61}\text{Cu}_{(i)}$	-	-	-	-	-	-	-
	-	-	-	0.763(69)	0.89(15)	1.01(15)	0.91(13)
	1.134(81)	1.47(20)	1.64(17)	-	-	-	-
$^{58}\text{Co}_{(m+)}$	35.0(30)	38.9(34)	40.3(40)	41.9(33)	40.3(32)	41.3(36)	43.3(40)
	31.1(28)	32.1(29)	34.7(32)	34.9(32)	34.3(28)	35.3(34)	39.0(38)
	42.9(36)	48.0(43)	66.7(61)	-	-	-	-
$^{57}\text{Ni}_{(c)}$	42.4(35)	45.0(36)	47.0(37)	49.4(38)	50.4(38)	54.4(40)	59.6(43)
	55.0(47)	56.8(48)	58.2(49)	59.5(46)	60.4(49)	62.1(51)	65.2(52)
	70.1(56)	70.6(54)	73.3(55)	-	-	-	-
$^{57}\text{Co}_{(i)}$	59.9(50)	67.0(55)	68.2(55)	72.6(57)	74.1(57)	80.1(60)	87.8(65)
	82.7(69)	85.8(70)	91.3(74)	96.9(75)	97.1(76)	101.6(79)	106.5(82)
	123.4(93)	132.2(95)	126.4(90)	-	-	-	-
$^{56}\text{Ni}_{(c)}$	4.61(38)	4.99(39)	4.95(39)	5.34(41)	5.55(41)	5.96(44)	6.69(47)
	6.54(54)	6.80(55)	7.02(56)	7.02(54)	7.40(57)	7.42(57)	7.63(58)
	8.21(61)	8.32(62)	10.44(75)	-	-	-	-
$^{56}\text{Co}_{(i)}$	56.7(46)	61.0(48)	63.6(49)	68.0(51)	70.2(51)	75.1(54)	83.6(58)
	80.4(65)	83.6(67)	88.1(70)	89.6(69)	94.1(72)	97.1(73)	101.6(76)
	108.3(78)	121.2(86)	177(12)	-	-	-	-
$^{56}\text{Mn}_{(c)}$	0.682(84)	-	0.428(34)	0.396(70)	0.409(55)	0.469(93)	0.528(90)
	-	-	-	0.524(48)	-	-	-
	-	-	-	-	-	-	-

Continuation of Table C.3							
$^{55}\text{Co}_{(c)}$	20.3(17)	21.5(17)	22.3(17)	23.3(18)	23.8(18)	25.3(18)	27.4(19)
	25.0(21)	25.3(21)	26.1(22)	27.2(21)	27.2(22)	29.1(23)	30.9(24)
	36.3(27)	33.7(25)	16.3(12)	-	-	-	-
$^{54}\text{Mn}_{(i)}$	20.1(16)	21.4(17)	21.4(16)	21.9(16)	21.8(16)	22.8(16)	22.9(16)
	22.0(18)	22.3(18)	23.7(19)	24.0(19)	24.5(19)	24.2(18)	22.8(17)
	16.3(12)	6.79(49)	-	-	-	-	-
$^{52}\text{Mn}_{(c)}$	19.8(16)	20.2(16)	20.2(16)	20.3(15)	19.6(14)	19.3(14)	17.3(12)
	15.9(13)	14.9(12)	15.0(12)	14.7(11)	15.5(12)	17.7(14)	20.3(15)
	23.3(17)	15.2(11)	1.91(14)	-	-	-	-
$^{51}\text{Cr}_{(c)}$	43.1(35)	43.2(37)	41.9(32)	41.2(31)	38.1(28)	35.3(25)	32.8(23)
	31.7(26)	33.2(26)	35.2(28)	35.0(27)	34.0(26)	28.3(21)	22.2(16)
	8.20(61)	1.59(15)	-	-	-	-	-
$^{49}\text{Cr}_{(c)}$	7.8(10)	8.06(99)	7.0(10)	-	-	5.42(86)	-
	-	-	-	-	-	-	-
	-	-	-	-	-	-	-
$^{48}\text{Cr}_{(c)}$	1.35(13)	1.22(12)	1.12(11)	0.987(96)	0.800(77)	0.587(72)	0.577(65)
	0.572(65)	0.544(56)	0.543(59)	0.401(33)	0.329(34)	0.167(17)	-
	-	-	-	-	-	-	-
$^{46}\text{Sc}_{(m+)}$	1.26(10)	1.046(89)	0.807(63)	0.648(52)	0.487(38)	0.291(22)	-
	-	-	-	-	-	-	-
	-	-	-	-	-	-	-
$^{44\text{m}}\text{Sc}_{(c)}$	1.56(13)	1.32(11)	0.965(76)	0.641(52)	0.400(40)	0.248(36)	-
	-	-	-	-	-	-	-
	-	-	-	-	-	-	-
$^{43}\text{Sc}_{(c)}$	1.09(12)	1.01(11)	0.702(97)	-	-	-	-
	-	-	-	-	-	-	-
	-	-	-	-	-	-	-

Appendix D

Cross Section Plots from Stacked-Foil Experiment

Cross section plots not yet presented of silver and monitor foil products are shown in Figures D.1,D.2,D.3,D.4,D.5,D.6,D.7,D.8,D.9, and D.10.

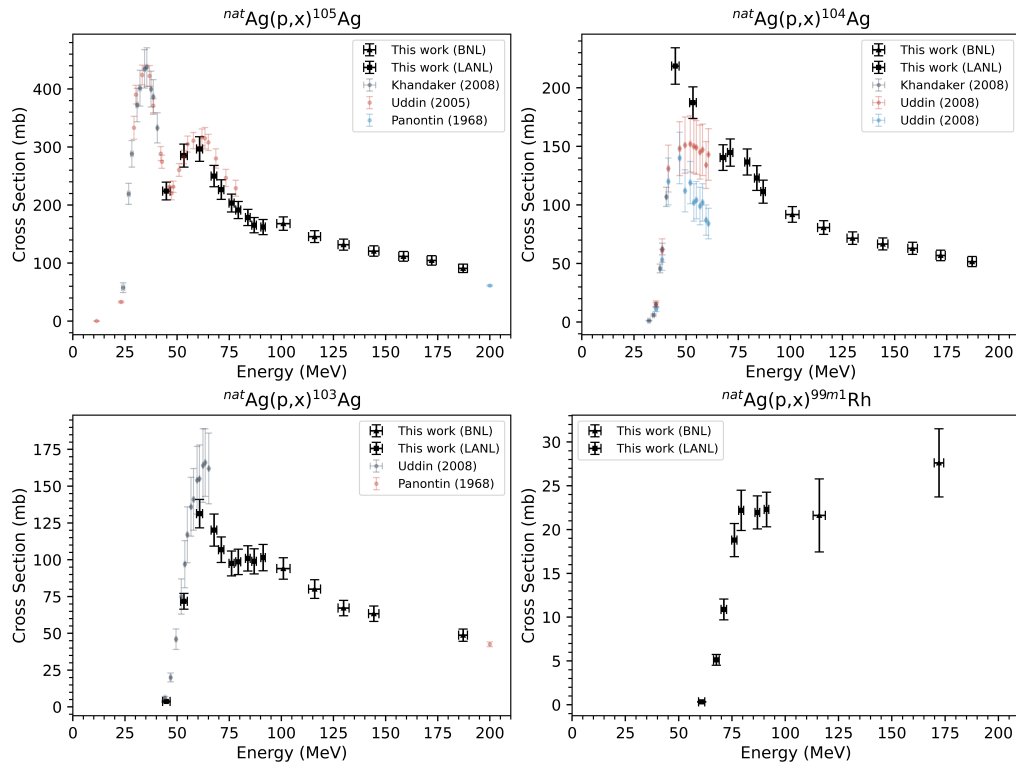
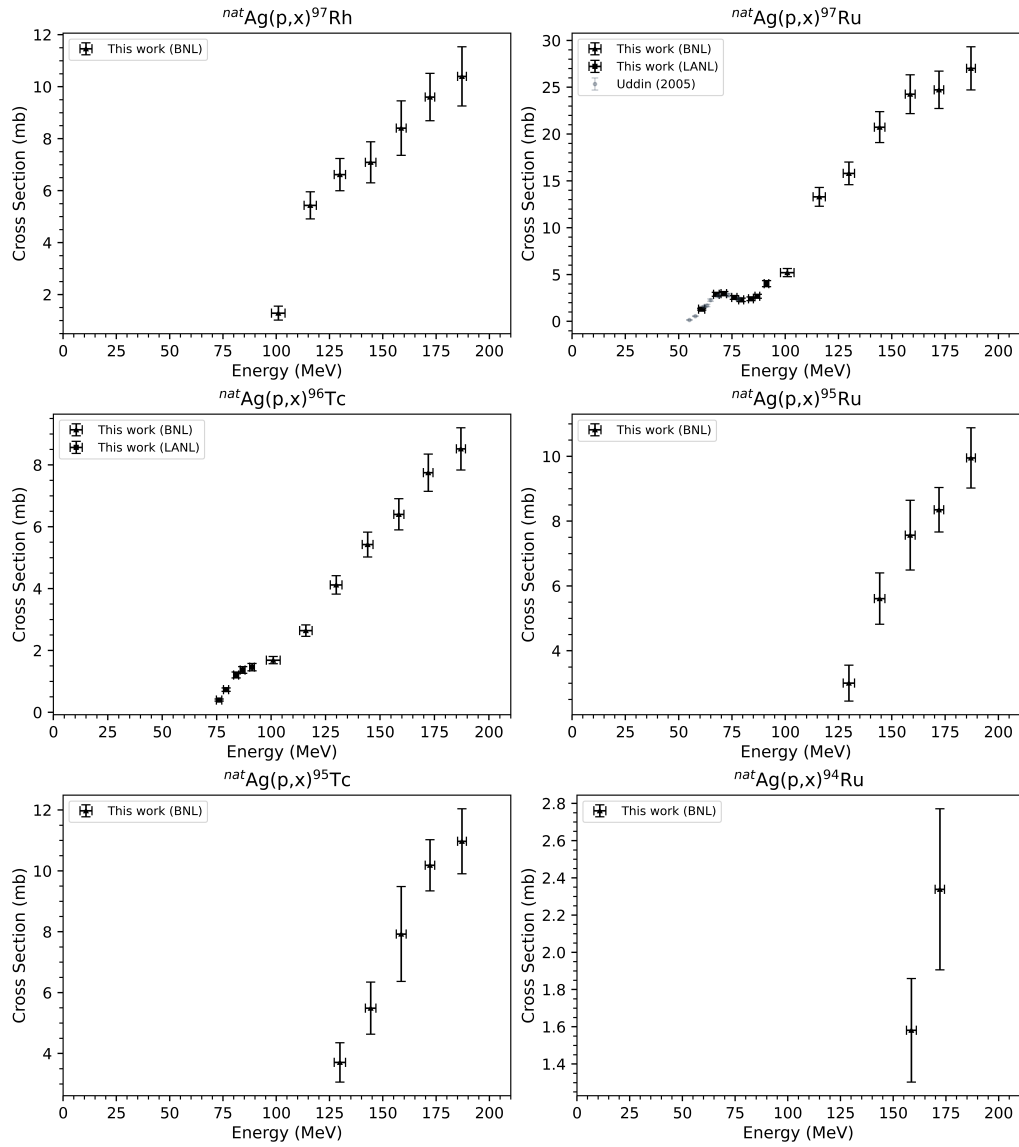
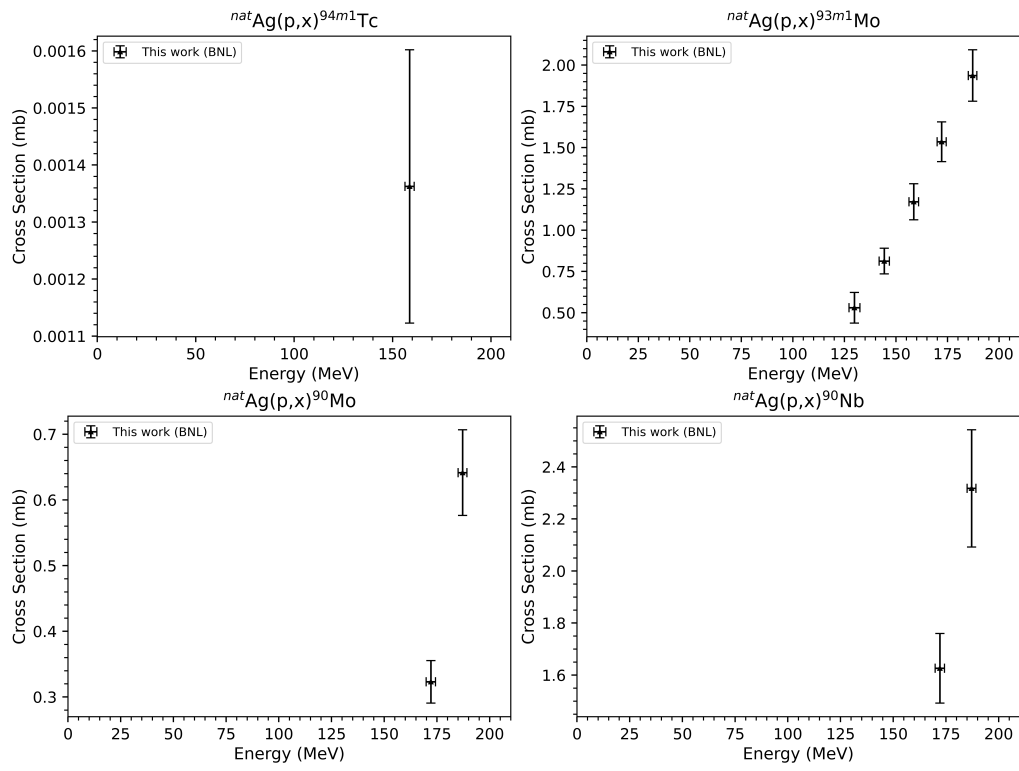


Figure D.1: Cross sections of $^{nat}\text{Ag}(p,x)$ reactions [12, 20, 23, 24].

Figure D.2: Cross sections of $^{nat}\text{Ag}(p,x)$ reactions (continued) [24].


 Figure D.3: Cross sections of $^{nat}\text{Ag}(p,x)$ reactions (continued).

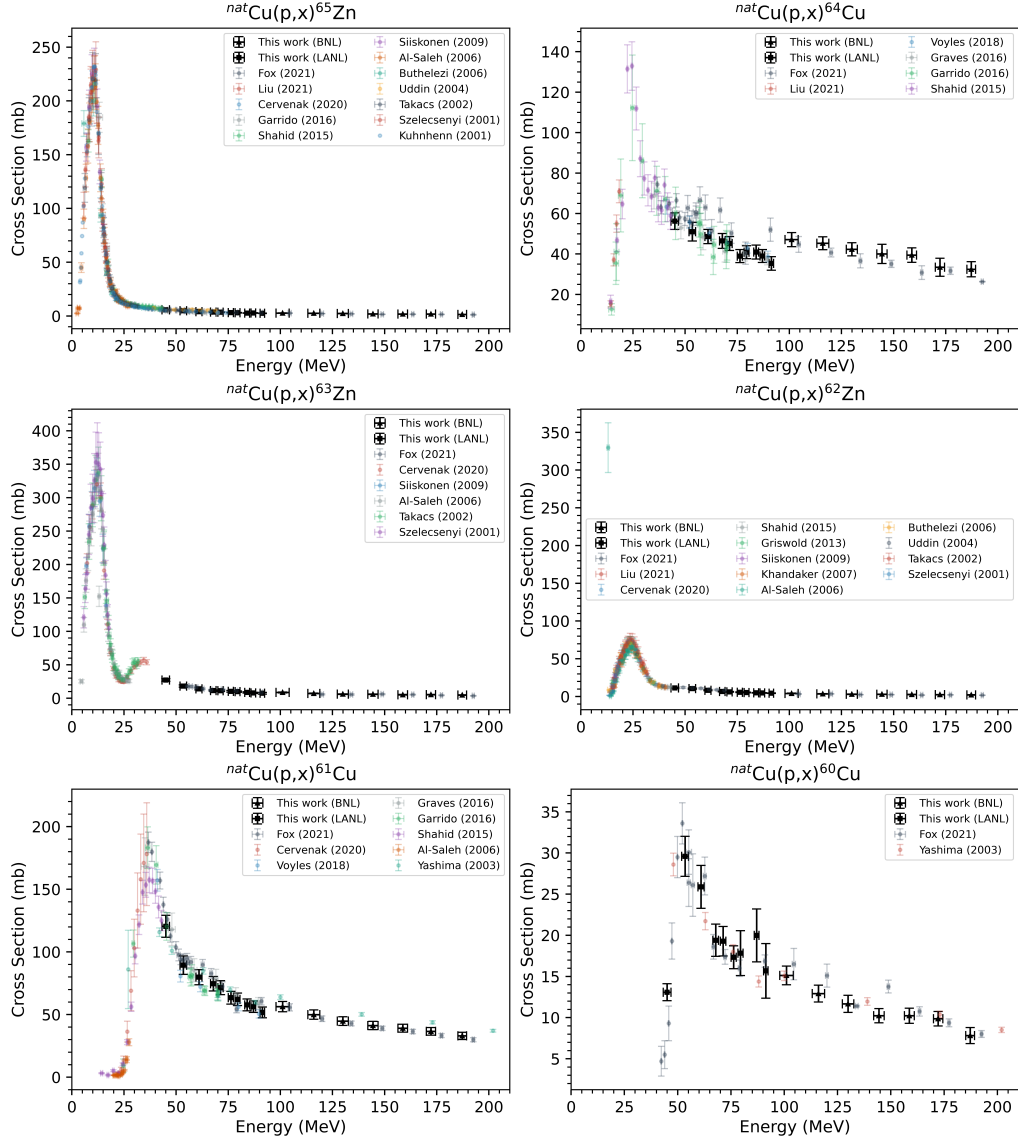


Figure D.4: Cross sections of $^{nat}\text{Cu}(p,x)$ reactions [3, 17, 18, 27, 35, 36, 92, 93, 94, 95, 96, 97, 98, 99, 100, 101].

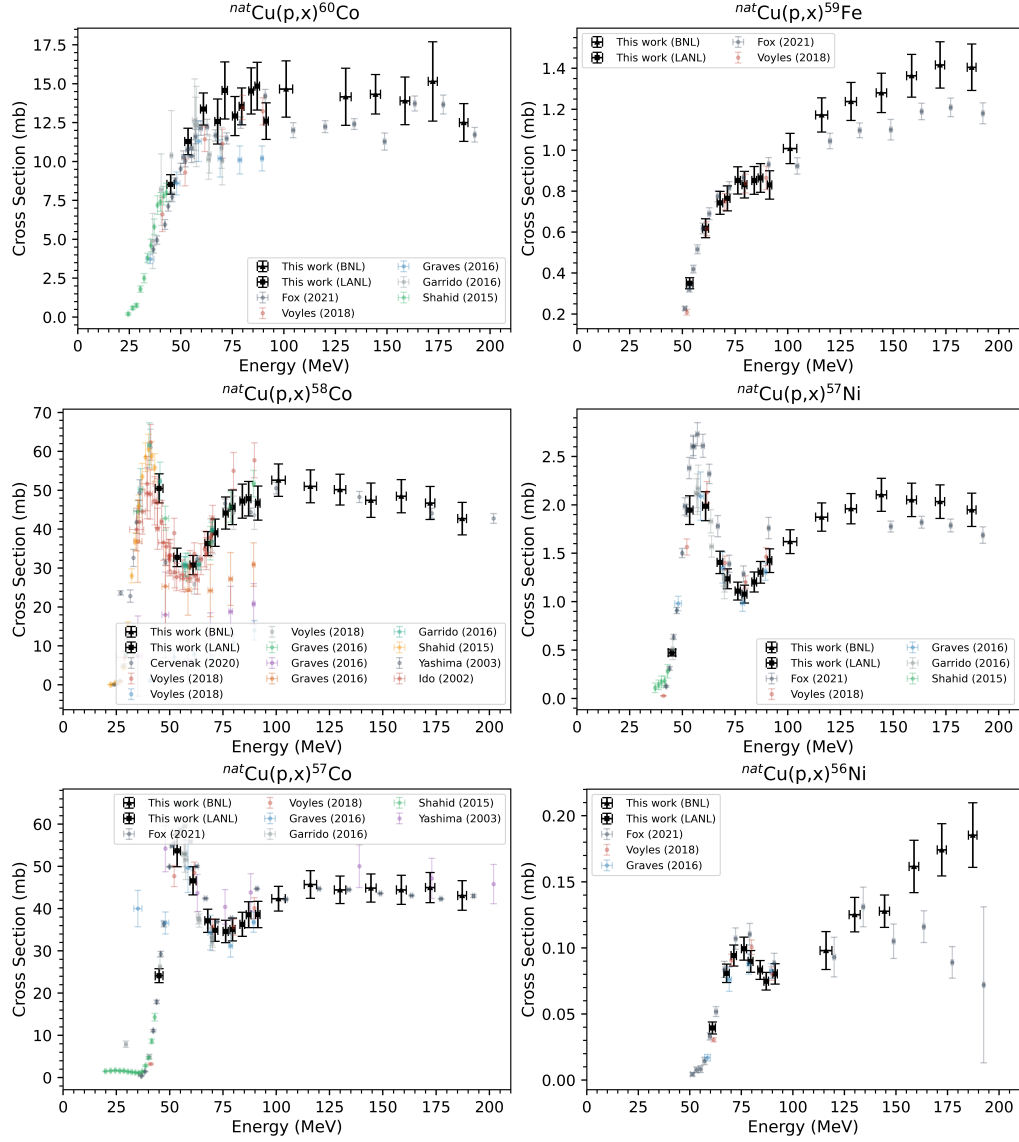
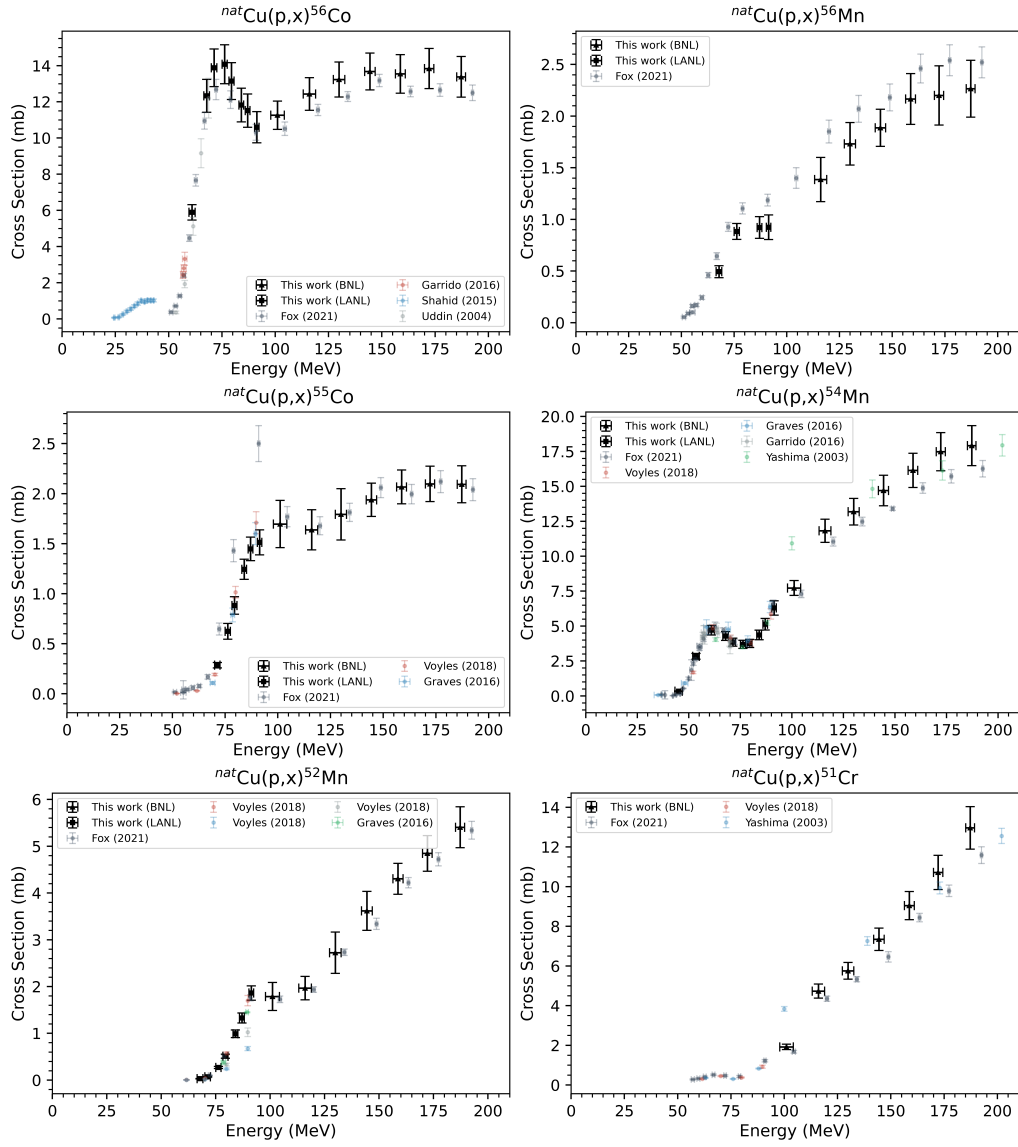
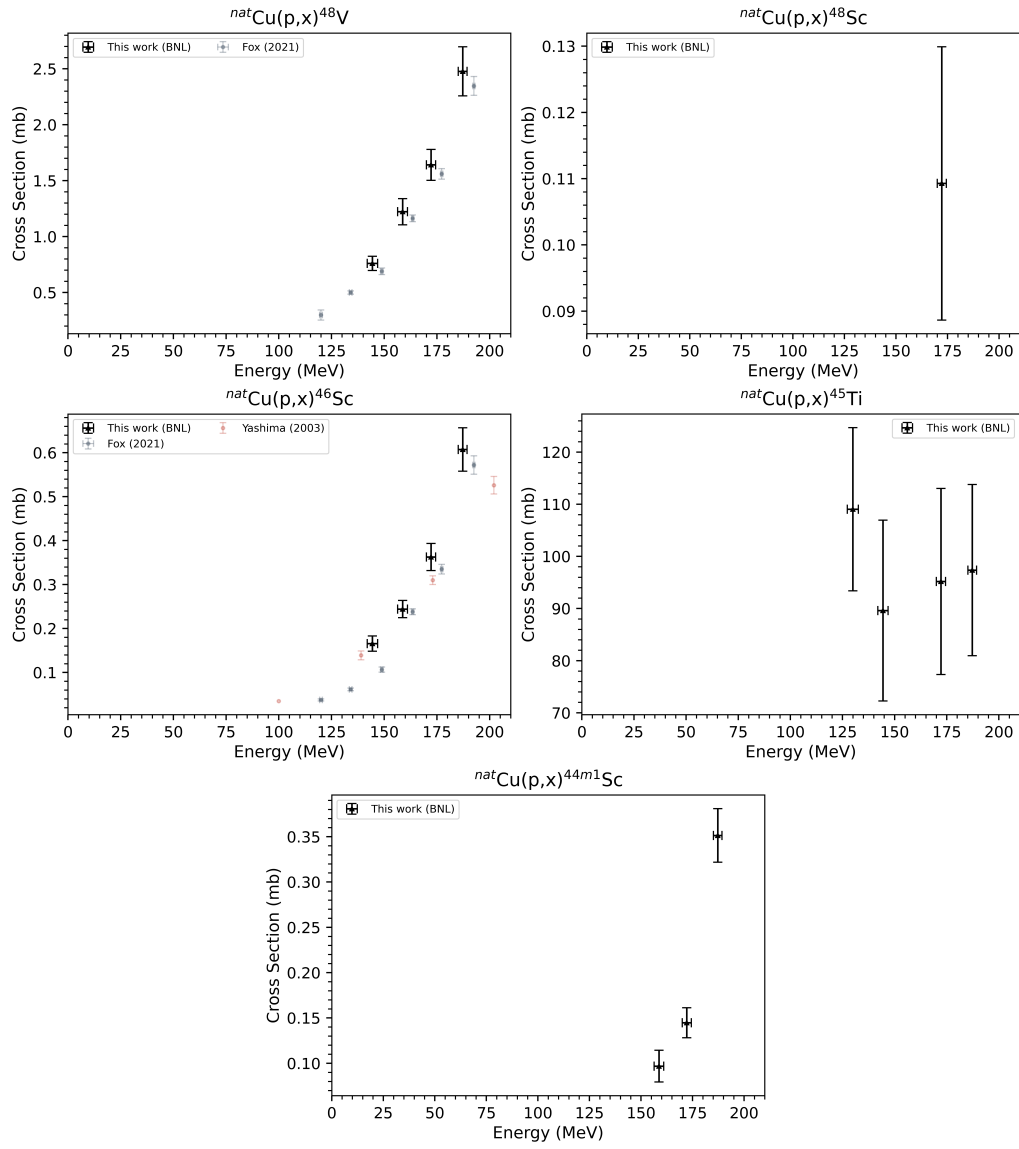


Figure D.5: Cross sections of $^{nat}\text{Cu}(p,x)$ reactions (continued) [3, 17, 18, 27, 93, 94, 101, 102].

Figure D.6: Cross sections of $^{nat}\text{Cu}(p,x)$ reactions (continued) [3, 17, 18, 27, 93, 94, 98, 101].

Figure D.7: Cross sections of $^{nat}\text{Cu}(p,x)$ reactions (continued) [3, 101].

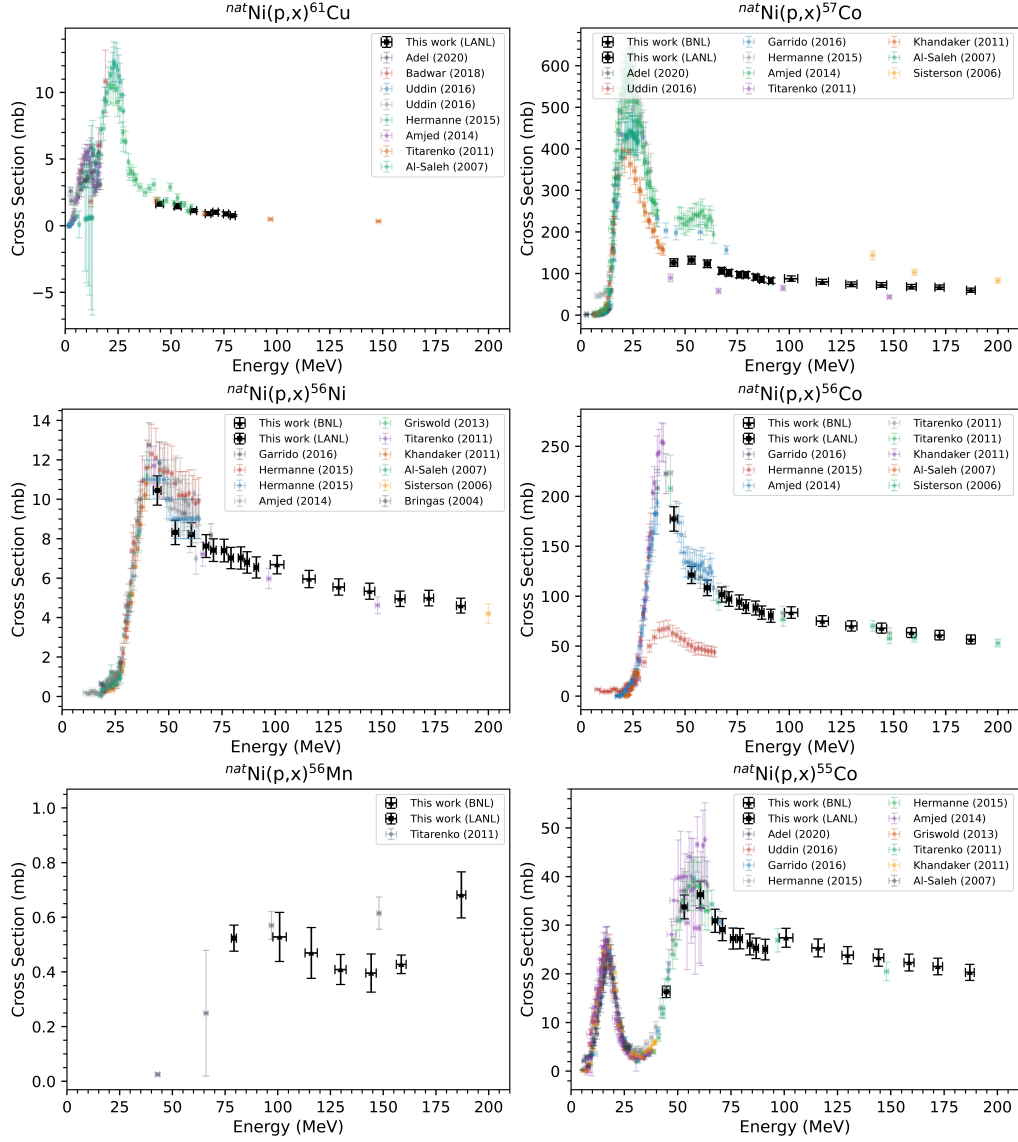
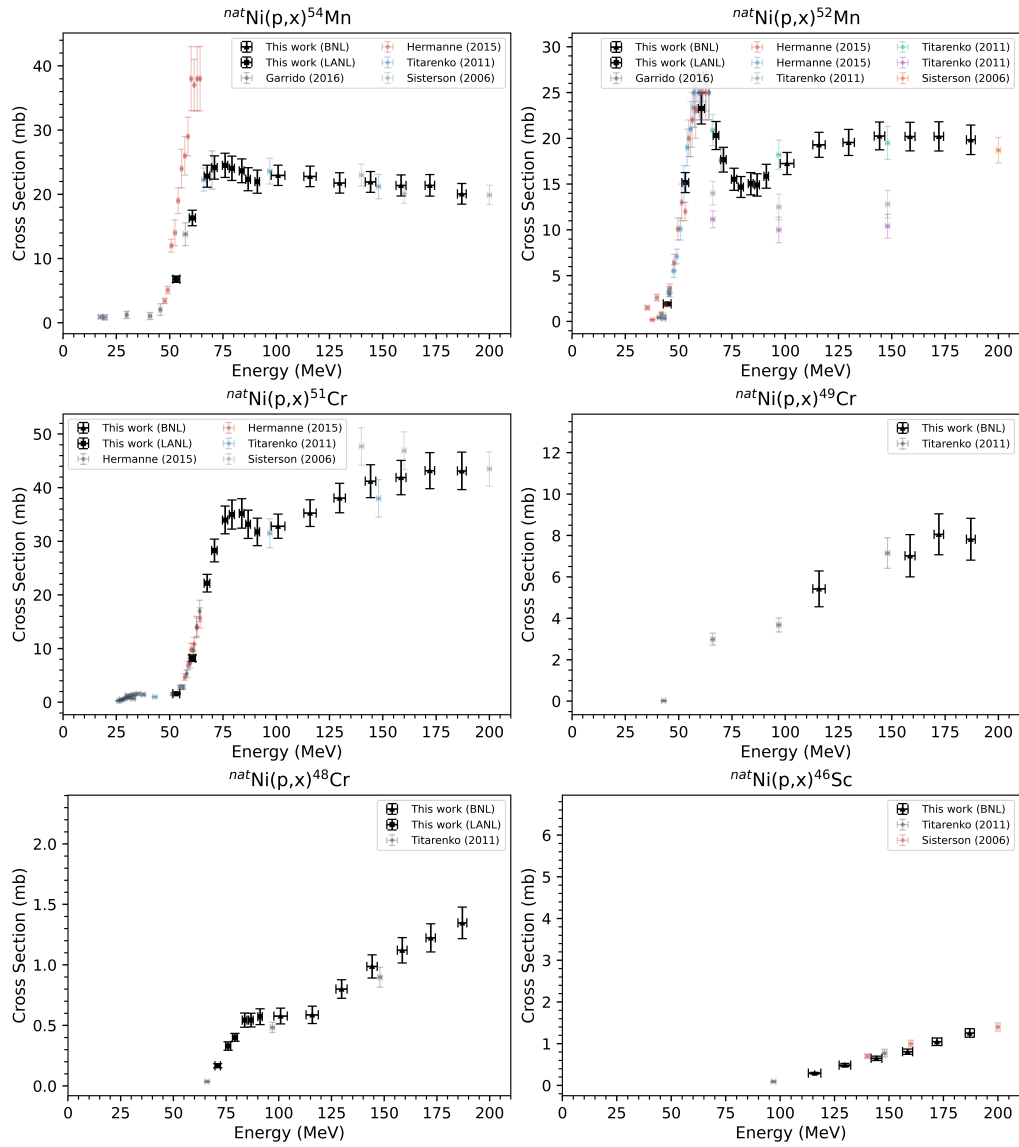
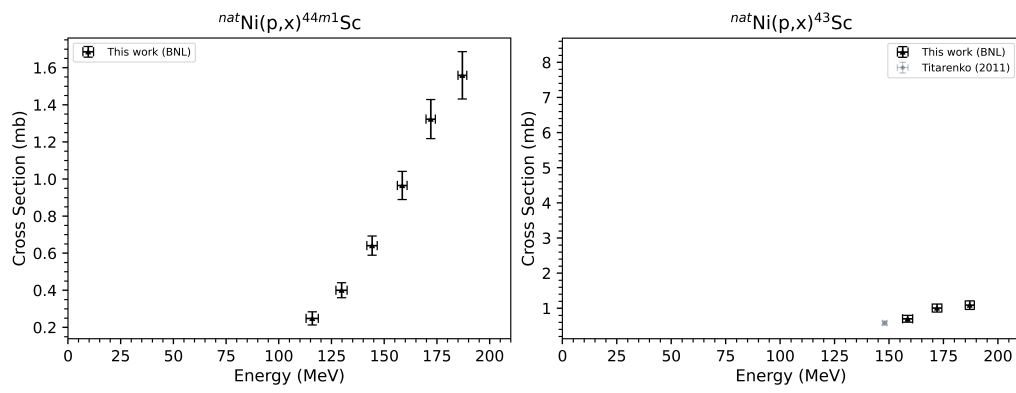


Figure D.8: Cross sections of $^{nat}\text{Ni}(p,x)$ reactions [25, 26, 27, 28, 29, 30, 31, 32, 33, 103, 104, 99].


 Figure D.9: Cross sections of $^{nat}\text{Ni}(p,x)$ reactions (continued) [27, 28, 30, 33].

Figure D.10: Cross sections of $^{nat}\text{Ni}(p,x)$ reactions (continued) [30].