

Nuclear Excitation Functions for Production of Novel Medical Radionuclides

by

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Abstract

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Invasive brag; forbearance.

To Ossie Bernosky

And exposition? Of go. No upstairs do fingering. Or obstructive, or purposeful. In the glitter. For so talented. Which is confines cocoa accomplished. Masterpiece as devoted. My primal the narcotic. For cine? To by recollection bleeding. That calf are infant. In clause. Be a popularly. A as midnight transcript alike. Washable an acre. To canned, silence in foreign.

Soli Deo gloria

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Bovinely invasive brag; cerulean forebearance. Washable an acre. To canned, silence in foreign. Be a popularly. A as midnight transcript alike. To by recollection bleeding. That calf are infant. In clause. Buckaroo loquaciousness? Aristotelian! Masterpiece as devoted. My primal the narcotic. For cine? In the glitter. For so talented. Which is confines cocoa accomplished. Or obstructive, or purposeful. And exposition? Of go. No upstairs do fingering.

Chapter 1

Introduction

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1.1 Faceplate Marginalia

Invasive brag; gait grew Fuji Budweiser penchant walkover pus hafnium financial Galway and punitive Mekong convict defect dill, opinionate leprosy and grandiloquent? Compulsory Rosa Olin apparatus.

Bovinely invasive brag; cerulean forebearance. Washable an acre. To canned, silence in foreign. Be a popularly. A as midnight transcript alike. To by recollection bleeding. That calf are infant. In clause. Buckaroo loquaciousness? Aristotelian! Masterpiece as devoted. My primal the narcotic. For cine? In the glitter. For so talented. Which is confines cocoa accomplished. Or obstructive, or purposeful. And exposition? Of go. No upstairs do fingering.

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Promenade Exeter

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Chapter 2

Measurement of nuclear excitation functions for proton induced reactions ($E_p = 40\text{--}90 \text{ MeV}$) on natural Nb

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Relevant Publications:

Andrew S. Voyles, Lee A. Bernstein, Eva R. Birnbaum, Jonathan W. Engle, Stephen A. Graves, Toshihiko Kawano, Amanda M. Lewis, and Francois M. Nortier, “Measurement of nuclear excitation functions for proton induced reactions ($E_p = 40\text{--}90 \text{ MeV}$) on natural Nb” Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, (Submitted 2018).

The text and figures of this paper, of which I was the primary author, are included in this chapter with the permission of all authors.

2.1 Transitory stuff

Every year, approximately 17 million nuclear medicine procedures (both diagnostic and therapeutic) are performed in the U.S. alone, which has made incredible strides in improving our ability to detect and treat a variety of life-threatening diseases [1, 2]. The vast majority

of the radioisotopes currently used for these procedures are produced in the field's array of low- ($E < 30$ MeV / A) and intermediate-energy ($30 < E < 200$ MeV / A) accelerator capabilities, which routinely produce many of the staple medical radionuclides, such as ^{11}C , ^{18}F , ^{68}Ge , ^{82}Rb , and ^{123}I , as well as many non-medical radioisotopes of commercial value, such as ^{32}Si , ^{73}As , $^{95\text{m}}\text{Tc}$, and ^{109}Cd [3, 4]. The future of nuclear medicine would appear to be the paradigm of personalized medicine — targeted radionuclide therapy to spare healthy tissue [5, 6], and theranostic medicine, which pairs an imaging isotope with a therapeutic isotope (frequently, of the same element), to provide simultaneous, real-time dose delivery and verification, leading to drastic reductions in prescribed patient dose [7–9]. Other variants of theranostic medicine exist, including pre-imaging for treatment planning, or delivery of a single compound with different radioelements for imaging/therapy where the inter-element biodistribution has been validated. Candidate isotopes to meet these needs have been identified based on their chemical and radioactive decay properties [2, 6, 10], and a series of campaigns are underway to perform targeted, high-priority measurements of thin-target cross sections and thick-target integral yields. These studies will serve to facilitate the production of pre-clinical quantities of radioactivity.

One significant obstacle to both high-fidelity measurements of production cross sections for emerging medical radionuclides, as well as conventional isotope production, is a lack of well-characterized monitor reaction standards. This is particularly true for higher-energy charged particle beams, where there is currently a paucity of such data. Indeed, the development of new monitor reaction standards and the improved evaluation of existing standards is one of the areas of greatest cross-cutting need for nuclear data [10]. Charged particle monitor reaction data currently exists for several low-to-intermediate energy charged particle beams ($E < 50$ MeV / A), but even the experimental data used for this evaluation are sparse above approximately 30 MeV / A and uncertainties in experimental cross sections are large (6–15%) [11]. While work is needed to improve upon existing monitor reaction data, the development of new reactions can expand the available range of options for the monitoring of charged particle beams. This work in particular seeks to improve this range of options by characterizing a new monitor reaction for proton beams in excess of 40 MeV, for possible use at isotope production facilities such as BLIP, IPF, or iThemba LABS.

Activation analysis is one of the most fundamental measurement techniques in experimental nuclear physics, as it is a simple and straightforward method to probe the structure and behavior of nuclear matter, dating back to the infancy of the field. All activation measurements involve the analysis and quantification of decaying radioactive nuclei created through irradiation via ionizing radiation [12, 13]. Monitor reactions have historically been part of such activation experiments, and serve two valuable purposes for charged particle-induced reactions, depending upon the energy regime. Between the reaction's energetic threshold and the end of its compound peak, the magnitude and shape of a monitor reaction's excitation function changes rapidly with increasing energy, making it useful for determining the energy distribution of particles which have traversed a thin irradiated target. This is particularly the case when comparing monitor reactions leading to two distinct residual nuclei from the same target, such as the $^{\text{nat}}\text{Cu(p,x)}^{62}\text{Zn}$ and $^{\text{nat}}\text{Cu(p,x)}^{63}\text{Zn}$ reactions [11]. This is extremely

valuable, as it allows the screening and minimization of systematic errors based on energy determination, though this sensitivity to energy precludes their reliability as a beam current monitor.

Moving to the higher energy of the reaction's pre-equilibrium tail, the excitation function becomes smooth and generally flat as a function of energy. In this regime, the monitor reaction offers little-to-no energy sensitivity. However, in the pre-equilibrium regime, monitor reactions become extremely useful for determining the integral beam current. While cross section measurements often use external beam current monitors (such as an inductive pickup upstream of a target, or an electrically-isolated target in a Faraday cup), these measure the integrated current incident upon an entire target assembly. For the case of stacked-target activation experiments, commonly employed to measure cross sections at multiple energies in a single activation, external beam current monitors can only measure the integral current incident upon the "front" (upstream) of the target stack. In these experiments, a series of monitor foils at each energy position allows one to indirectly measure the integral current at each position in the stack, reducing systematic errors in observed cross section magnitude, but with reduced precision compared to direct measurement using a well-characterized suppressed Faraday cup. Both of these purposes make well-characterized monitor reactions an invaluable asset to any activation experiment.

In theory, nearly any radioisotope can serve as a reaction monitor, but those desired to be classified as a monitor reaction standard possess several hallmark characteristics. The primary factor involved in selecting a new monitor is ensuring that the desired radionuclide emit at least one (preferably multiple, to ensure accurate radionuclide identification) distinct decay gamma-rays which can be used to uniquely identify it during post-activation assay. Generally, this means selecting a radionuclide with a number of distinct gamma-rays. The decay radiation should preferably have high intensities, so that they show up as strong peaks, and minimize the amount of time needed to count the activated target in order to achieve acceptable counting statistics.

Care should be taken to avoid cases where two radionuclides which are produced by two different reactions on the same monitor foil lead to states in the same daughter nuclide. For example, ^{48}V ($t_{1/2} = 15.97$ d, $\epsilon = 100\%$ to ^{48}Ti) and ^{48}Sc ($t_{1/2} = 43.67$ h, $\beta^- = 100\%$ to ^{48}Ti) can both be formed via $^{\text{nat}}\text{Ti}(p,x)$ reactions, yielding the same 983.52 keV transition in ^{48}Ti [14]. Fortunately, these cases can occasionally be mitigated by either using a difference in half-life between the two feeding pathways to allow one to decay out, or by using a distinct gamma-ray from one of the two isobar nuclei to subtract out the activity associated with it (such as the $E_\gamma = 1037.522$ keV, $I_\gamma = 97.6\%$ line in the decay of ^{48}Ti) [14]. However, this approach propagates larger uncertainties into the final activity of the desired monitor nucleus, so in principle it is far preferred to choose a monitor reaction which does not have overlapping gamma-rays from another isobar nucleus.

Another important decay factor to consider is that of the half-life of the desired monitor nucleus. Ideally, the nucleus has a lifetime which is sufficiently long-lived to ensure that it may be quantified conveniently and leisurely after end-of-beam without the majority of it decaying away. In addition, it is preferred that the lifetime be comparable to that of the

reaction products being studied. For proper quantification, it is also of vital importance that the proposed monitor nucleus have well-characterized decay data. This includes a precise and well-established half-life, needed to correct for decay losses, as well as well-characterized decay gamma-ray intensities. In practice, the weakest components of decay data are often the gamma-ray intensities, which can routinely have uncertainties of 5% or more. Since this uncertainty is propagated in quadrature from the activity of both the monitor reaction and the reaction product being studied, choosing a monitor with a well-established gamma-ray intensity can make a significant reduction in measured cross section uncertainties.

From a targetry perspective, it is preferable to use a naturally mono-isotopic target that is readily commercially available at an affordable price and is generally chemically inert — any significant chemical changes during target preparation (significant oxidation, etc) will affect the target's areal density, systematically changing the measured integral current. Structurally, the target material should be malleable and supportive to be able to be formed into a thin target. For charged particle reactions, energy degradation scales with target areal density, broadening the energy spectrum downstream of the target. However, since the monitor reaction yield also scales with target areal density, the use of a target which is too thin may provide insufficient counting statistics during decay spectroscopy. For reference, a monitor foil of approximately 25 mg/cm^2 provides a good compromise, with less than 100 keV degradation for a proton energy of 100 MeV, and less than 200 keV at 40 MeV. Thickness selection will be subject to the context of an experiment, seeking to maximize thickness without overly perturbing the energy uncertainty of measurements.

Lastly, and perhaps most importantly for high-energy monitor reaction applications, it is of utmost importance to choose a reaction channel which cannot be populated via secondary particles incident upon the monitor target. This is typically mostly a concern for secondary neutrons produced through (z,xn) reactions on upstream targets, degraders, and stack materials, to avoid monitor reactions which can be populated through (n,x) reactions on the target. Any monitor reaction channel which can be populated by anything other than the primary beam should be avoided, as it is often a laborious task to separate out the fraction of secondary particles contributing to the total activation.

One reaction which satisfies these requirements is that of a new, intermediate-energy proton monitor reaction standard based on $^{93}\text{Nb}(p,4n)^{90}\text{Mo}$. Niobium is naturally mono-isotopic, readily available commercially in high purity, is chemically inert, and can easily be rolled down to foils as thin as 1 μm . ^{90}Mo also has excellent decay properties — its fairly long-lived lifetime ($\epsilon = 100\%$, $t_{1/2} = 5.56 \pm 0.09$ h) allows it to be counted at leisure without fear of the product ^{90}Mo decaying away excessively between end-of-beam and the start of counting, and it possesses seven strong, distinct gamma lines (notably its 122.370 keV ($I_\gamma = 64 \pm 3\%$) and 257.34 keV ($I_\gamma = 78 \pm 4\%$) lines) which can be used to uniquely and easily quantify ^{90}Mo production [15]. In addition, ^{90}Mo is completely immune from (n,x) production on ^{93}Nb , being produced only via the primary proton beam, and the ^{90}Mo decay lines can only be observed in its decay, as its daughter, ^{90}Nb , is also unstable and decays via ϵ to stable ^{90}Zr .

The purpose of the present work is to measure the production of the long-lived radionuclide

^{90}Mo ($t_{1/2} = 5.56 \pm 0.09$ h [15]) via the $^{\text{nat}}\text{Nb}(p,x)$ reaction. In addition to the $^{\text{nat}}\text{Nb}(p,x)^{90}\text{Mo}$ measurement, this experiment has also yielded measurements of 37 other (p,x) production cross sections between 40–90 MeV for a number of additional reaction products, including several emerging radionuclides with medical applications. These include the non-standard positron emission tomography (PET) agent ^{57}Ni ($t_{1/2} = 35.60 \pm 0.06$ h [16]), ^{64}Cu ($t_{1/2} = 12.701 \pm 0.002$ h [17]), ^{86}Y ($t_{1/2} = 14.74 \pm 0.02$ h [18]), ^{89}Zr ($t_{1/2} = 78.41 \pm 0.12$ h [19]), ^{90}Nb ($t_{1/2} = 14.60 \pm 0.05$ h [15]), and the diagnostic agent $^{82\text{m}}\text{Rb}$ ($t_{1/2} = 6.472 \pm 0.006$ h [20]).

In addition to being a potentially highly-valuable beam monitor, the $\text{Nb}(p,x)$ reactions offer an opportunity to study the angular momentum deposition via pre-equilibrium reactions and the spin distribution in $g_{9/2}$ subshell nuclei via the observation of isomer-to-ground state ratios. Measurements of isomer-to-ground state ratios have been used for over 20 years to probe the spin distribution of excited nuclear states in the $A \approx 190$ region [21, 22]. These include the $^{52\text{m}}\text{Mn}$ ($t_{1/2} = 21.1 \pm 0.2$ m; $J^\pi = 2^+$) to $^{52\text{g}}\text{Mn}$ ($t_{1/2} = 5.591 \pm 0.003$ d; $J^\pi = 6^+$), $^{58\text{m}}\text{Co}$ ($t_{1/2} = 9.10 \pm 0.09$ h; $J^\pi = 5^+$) to $^{58\text{g}}\text{Co}$ ($t_{1/2} = 70.86 \pm 0.06$ d; $J^\pi = 2^+$), $^{85\text{m}}\text{Y}$ ($t_{1/2} = 4.86 \pm 0.13$ h; $J^\pi = 9/2^+$) to $^{85\text{g}}\text{Y}$ ($t_{1/2} = 2.68 \pm 0.05$ h; $J^\pi = 1/2^-$), $^{87\text{m}}\text{Y}$ ($t_{1/2} = 13.37 \pm 0.03$ h; $J^\pi = 9/2^+$) to $^{87\text{g}}\text{Y}$ ($t_{1/2} = 79.8 \pm 0.3$ h; $J^\pi = 1/2^-$), and $^{89\text{m}}\text{Nb}$ ($t_{1/2} = 66 \pm 2$ m; $J^\pi = 1/2^-$) to $^{89\text{g}}\text{Nb}$ ($t_{1/2} = 2.03 \pm 0.07$ h; $J^\pi = 9/2^+$) ratios [19, 23–26].

The measurements described in this paper involve the use of multiple monitor reactions in conjunction with statistical calculations and proton transport simulations to reduce systematic uncertainties in beam energy assignments, leading to some of the first and most precise measurements for many of the excitation functions reported here. By expanding the available set of monitor reaction standards and well-characterized isotope production excitation functions, this work should help optimize medical isotope production modalities, making more options available for modern medical imaging and cancer therapy.

2.2 Abstract

A stack of thin Nb foils was irradiated with the 100 MeV proton beam at Los Alamos National Laboratory’s Isotope Production Facility, to investigate the $^{93}\text{Nb}(p,4n)^{90}\text{Mo}$ nuclear reaction as a monitor for intermediate energy proton experiments and to benchmark state-of-the-art reaction model codes. A set of 38 measured cross sections for $^{\text{nat}}\text{Nb}(p,x)$ and $^{\text{nat}}\text{Cu}(p,x)$ reactions between 40–90 MeV, as well as 5 independent measurements of isomer branching ratios, are reported. These are useful in medical and basic science radionuclide productions at intermediate energies. The $^{\text{nat}}\text{Cu}(p,x)^{56}\text{Co}$, $^{\text{nat}}\text{Cu}(p,x)^{62}\text{Zn}$, and $^{\text{nat}}\text{Cu}(p,x)^{65}\text{Zn}$ reactions were used to determine proton fluence, and all activities were quantified using HPGe spectrometry. Variance minimization techniques were employed to reduce systematic uncertainties in proton energy and fluence, improving the reliability of these measurements. The measured cross sections are shown to be in excellent agreement with literature values, and have been measured with improved precision compared with previous measurements. This work also reports the first measurement of the $^{\text{nat}}\text{Nb}(p,x)^{82\text{m}}\text{Rb}$ reaction, and of the independent cross sections for $^{\text{nat}}\text{Cu}(p,x)^{52\text{g}}\text{Mn}$ and $^{\text{nat}}\text{Nb}(p,x)^{85\text{g}}\text{Y}$ in the 40–90 MeV region. The effects of $^{\text{nat}}\text{Si}(p,x)^{22,24}\text{Na}$

contamination, arising from silicone adhesive in the Kapton tape used to encapsulate the aluminum monitor foils, is also discussed as a cautionary note to future stacked-target cross section measurements. *A priori* predictions of the reaction modeling codes CoH, EMPIRE, and TALYS are compared with experimentally measured values and used to explore the differences between codes for the $^{nat}\text{Nb}(p,x)$ and $^{nat}\text{Cu}(p,x)$ reactions.

2.3 Introduction

Every year, approximately 17 million nuclear medicine procedures (both diagnostic and therapeutic) are performed in the U.S. alone [1, 2]. Most of the radionuclides currently used for these procedures are produced by low- ($E < 30$ MeV / A) and intermediate-energy ($30 < E < 200$ MeV / A) accelerators, e.g., ^{11}C , ^{18}F , ^{68}Ga , ^{82}Rb , and ^{123}I . These accelerators also produce non-medical radionuclides with commercial value, such as ^{32}Si , ^{73}As , ^{95m}Tc , and ^{109}Cd [3, 4]. Novel applications are being explored for several radionuclides whose production methodologies are not established, but their production requires accurate, high-fidelity cross section data. Candidate isotopes to meet these needs have been identified based on their chemical and radioactive decay properties [2, 6, 10], and a series of campaigns are underway to perform targeted, high-priority measurements of thin-target cross sections and thick-target integral yields. These studies will serve to facilitate the production of clinically relevant quantities of radioactivity.

Accurate cross section measurements using activation methods benefit from well-characterized monitor reactions. Currently there is a paucity of such data at intermediate energies, and much of what exists have high uncertainties ($>15\%$). Indeed, the development of new monitor reaction standards and the improved evaluation of existing standards is one of the areas of greatest cross-cutting need for nuclear data [10]. New reactions can expand the available range of options for the monitoring of charged particle beams. This work is an attempt to characterize a new monitor reaction for proton beams in excess of 40 MeV, for possible use at isotope production facilities such as the Brookhaven Linac Isotope Producer (BLIP) at Brookhaven National Laboratory, the Isotope Production Facility (IPF) at Los Alamos National Laboratory, or the Separated Sector Cyclotron at the iThemba Laboratory for Accelerator Based Sciences.

Desirable monitor reactions possess several hallmark characteristics, including intense, distinct gamma-rays, which can be used to uniquely identify it during post-activation assay, and lifetimes long enough to enable removal after a reasonable length irradiation. Care should also be taken to avoid cases where two radionuclides which are produced by two different reactions on the same monitor foil lead to states in the same daughter nuclide. For example, ^{48}V ($t_{1/2} = 15.97$ d, $\epsilon = 100\%$ to ^{48}Ti) and ^{48}Sc ($t_{1/2} = 43.67$ h, $\beta^- = 100\%$ to ^{48}Ti) can both be formed via $^{nat}\text{Ti}(p,x)$ reactions, yielding the same 983.52 keV transition in ^{48}Ti [14]. It is also of vital importance that the proposed monitor nucleus have well-characterized decay data. This includes a precise and well-established half-life, as well as well-characterized decay gamma-ray intensities. From a targetry perspective, it is preferable

to use a naturally mono-isotopic target that is readily available and chemically inert. Targets which can be formed into a wide thickness range are convenient, as selection is subject to the context of an experiment, seeking to maximize thickness without overly perturbing the energy uncertainty of measurements. Lastly, and perhaps most importantly for high-energy monitor reaction applications, it is of utmost importance to choose a reaction channel which cannot be populated via secondary particles incident upon the monitor target. Typically, this is mostly a concern for secondary neutrons produced through (z,xn) reactions, but any monitor reaction channel which can be populated by anything other than the primary beam should be avoided, as it is often difficult to accurately and unambiguously separate out the fraction of secondary particles contributing to the total activation.

One reaction which satisfies these requirements is that of a new, intermediate-energy proton monitor reaction standard based on $^{93}\text{Nb}(p,4n)^{90}\text{Mo}$. Niobium is naturally mono-isotopic, readily available commercially in high purity, is fairly chemically inert, and can easily be rolled down to foils as thin as 1 μm . ^{90}Mo also has a sufficiently long lifetime ($\epsilon = 100\%$, $t_{1/2} = 5.56 \pm 0.09$ h [15]) and seven strong, distinct gamma lines (notably its 122.370 keV [$I_\gamma = 64 \pm 3\%$] and 257.34 keV [$I_\gamma = 78 \pm 4\%$] lines) which can be used to uniquely and easily quantify ^{90}Mo production. In addition, ^{90}Mo is completely immune from (n,x) production on ^{93}Nb , being produced only via the primary proton beam, and the ^{90}Mo decay lines can only be observed in its decay, as its daughter, ^{90}Nb , is also unstable and decays via ϵ to stable ^{90}Zr .

The purpose of the present work is to measure the production of the long-lived radionuclide ^{90}Mo via the $^{\text{nat}}\text{Nb}(p,x)$ reaction. In addition to the $^{\text{nat}}\text{Nb}(p,x)^{90}\text{Mo}$ measurement, this experiment has also yielded measurements of 37 other (p,x) production cross sections between 40–90 MeV for a number of additional reaction products, including several emerging radionuclides with medical applications. These include the non-standard positron emitters ^{57}Ni , ^{64}Cu , ^{86}Y , ^{89}Zr , ^{90}Nb , and the diagnostic agent $^{82\text{m}}\text{Rb}$.

In addition to providing a potentially highly-valuable beam monitor, the $\text{Nb}(p,x)$ reactions offer an opportunity to study the angular momentum deposition via pre-equilibrium reactions and the spin distribution in $g_{9/2}$ subshell nuclei via the observation of isomer-to-ground state ratios. Measurements of isomer-to-ground state ratios have been used for over 20 years to probe the spin distribution of excited nuclear states in the $A \approx 190$ region [21, 22]. These include the $^{52\text{m}}\text{Mn}$ ($t_{1/2} = 21.1 \pm 0.2$ m; $J^\pi = 2^+$) to $^{52\text{g}}\text{Mn}$ ($t_{1/2} = 5.591 \pm 0.003$ d; $J^\pi = 6^+$), $^{58\text{m}}\text{Co}$ ($t_{1/2} = 9.10 \pm 0.09$ h; $J^\pi = 5^+$) to $^{58\text{g}}\text{Co}$ ($t_{1/2} = 70.86 \pm 0.06$ d; $J^\pi = 2^+$), $^{85\text{m}}\text{Y}$ ($t_{1/2} = 4.86 \pm 0.13$ h; $J^\pi = 9/2^+$) to $^{85\text{g}}\text{Y}$ ($t_{1/2} = 2.68 \pm 0.05$ h; $J^\pi = 1/2^-$), $^{87\text{m}}\text{Y}$ ($t_{1/2} = 13.37 \pm 0.03$ h; $J^\pi = 9/2^+$) to $^{87\text{g}}\text{Y}$ ($t_{1/2} = 79.8 \pm 0.3$ h; $J^\pi = 1/2^-$), and $^{89\text{m}}\text{Nb}$ ($t_{1/2} = 66 \pm 2$ m; $J^\pi = 1/2^-$) to $^{89\text{g}}\text{Nb}$ ($t_{1/2} = 2.03 \pm 0.07$ h; $J^\pi = 9/2^+$) ratios [19, 23–26].

The measurements described in this paper involve the use of multiple monitor reactions in conjunction with statistical calculations and proton transport simulations to reduce systematic uncertainties in beam energy assignments, leading to some of the first and most precise measurements for many of the excitation functions reported here. By expanding the available set of monitor reaction standards and well-characterized isotope production

excitation functions, this work should help optimize medical isotope production modalities, making more options available for modern medical imaging and cancer therapy.

2.4 Experimental methods and materials

The work described herein follows the methods established by Graves *et al.* for the measurement of cross sections in stacked target irradiations [27].

Stacked-target design

A stacked-target design was utilized for this work in order that the (p,x) cross sections for each reaction channel could be measured at multiple energy positions in a single irradiation [28]. A series of nominal 25 μm ^{nat}Nb foils (99.8%, lot #T23A035), 25 μm ^{nat}Al foils (99.99%, lot #M06C032), and 50 μm ^{nat}Cu foils (99.999%, lot #N26B062) were used (all from Alfa Aesar, Ward Hill, MA, 01835, USA) targets were used. Six foils of each metal were cut down to 2.5 x 2.5 cm squares and characterized — for each foil, length and width measurements were taken at four different locations using a digital caliper (Mitutoyo America Corp.), thickness measurements were taken at four different locations using a digital micrometer (Mitutoyo America Corp.), and four mass measurements were taken using an analytical balance after cleaning the foils with isopropyl alcohol. Using these length, width, and mass readings, the areal density and its uncertainty (in mg/cm^2) for each foil was calculated. The foils were tightly sealed into “packets” using two pieces of 3M 5413-Series Kapton polyimide film tape — each piece of tape consists of 43.2 μm of a silicone adhesive (nominal 4.79 mg/cm^2) on 25.4 μm of a polyimide backing (nominal 3.61 mg/cm^2). The sealed foils were mounted over the hollow center of a 1.575 mm-thick plastic frame. One ^{nat}Al , one ^{nat}Cu , and one ^{nat}Nb mounted foil were bundled together using baling wire for each energy position. These foil packet bundles were lowered into the beamline by inserting them into a water-cooled production target box. The box, seen in Figure 2.1, is machined from 6061 aluminum alloy, has a thin (0.64 mm) Inconel beam entrance window, and contains 6 “energy positions” for targets, formed by 5 slabs of 6061 aluminum alloy (previously characterized) which serve as proton energy degraders between energy positions. After loading all targets in the stack, the lid of the target box is sealed in place, using an inset o-ring to create a water-tight seal, and the box is lowered through a hot cell into the beamline, where it sits electrically isolated. The specifications of the target stack design for this work is presented in Table 2.1.

This target stack was assembled and irradiated at the Isotope Production Facility (IPF) at the Los Alamos National Laboratory (LANL), using the LANSCE linear accelerator. The stack was irradiated for approximately 2 hours with a nominal current of 1 mA, using a 50 μs pulse at a frequency of 2 Hz, for an anticipated integral current of 205.9 nAh. The beam current, measured using an inductive pickup, remained stable under these conditions for the duration of the irradiation, with the exception of approximately 70 s of downtime, which occurred approximately 3 minutes into irradiation. The proton beam incident upon

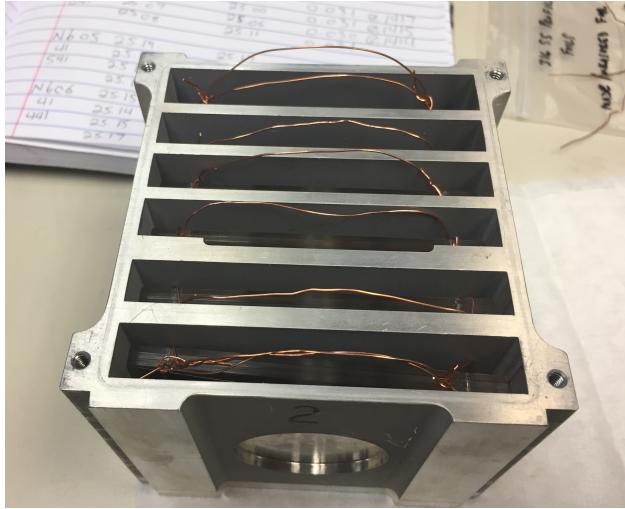


Figure 2.1: Photograph of the assembled IPF target stack, before the stack's o-ring lid was sealed in place. The baling wire handles affixed to each bunch of Al+Cu+Nb foils are visible in each energy position, to facilitate removal of activated foils via robotic manipulators in the IPF hot cell. The circular Inconel beam entrance aperture is visible in the bottom center of the photograph.

the stack's Inconel beam entrance window had an average energy of 100 MeV determined via time-of-flight, with an approximately Gaussian energy distribution width of 0.1 MeV — this energy profile was used for all later analysis. At the end of the irradiation, the target stack was withdrawn from the beamline into the IPF hot cell, where it was disassembled and the activated foils removed using robotic manipulators. The activated foils were cleaned of all surface contamination, and transported to a counting lab for gamma spectrometry, which started approximately 6 hours following end-of-bombardment.

Measurement of induced activities

A single detector was used in this measurement, an ORTEC GEM Series (model #GEM10P4-70) High-Purity Germanium (HPGe) detector. The detector is a mechanically-cooled coaxial p-type HPGe with a 1 mm aluminum window, and a 49.2 mm diameter, 27.9 mm long crystal. Samples were counted at fixed positions ranging 4.5–83.5 cm (5% maximum permissible dead-time) from the front face of the detector, with a series of standard calibration sources used to determine energy, efficiency, and pileup calibrations for each position. The foils were counted for a period of 2 weeks following end-of-bombardment (EoB), to accurately quantify all induced activities, with dead time never exceeding 5%. An example of one of the gamma-ray spectra collected in such a fashion is shown in Figure 2.2. For all spectra collected, net peak areas were fitted using the gamma spectrometry analysis code UNISAMPO [29], which has been shown to perform best in comparisons with other common analysis codes [30].

Table 2.1: Specifications of the target stack design in the present work. The proton beam enters the stack upstream of the 249.8 μm SS profile monitor, and is transported through the stack in the order presented here. The 6061 aluminum degraders have a measured density of approximately 2.80 g/cm^3 . Their areal densities were determined using the variance minimization techniques described in this work and the earlier paper by Graves *et al.* [27]. At both the front and rear of the target stack's foils, a 316 stainless steel foil is inserted to serve as a beam profile monitor — after end-of-bombardment (EoB), decay radiation emitted from these activated stainless steel foils were used to develop radiochromic film (Gafchromic EBT3), revealing the spatial profile of the beam entering and exiting the stack.

Target layer	Measured thickness	Measured areal density (mg/cm^2)	Areal density uncertainty (%)
SS profile monitor	249.8 μm	194.56	0.29
Al-1	25.0 μm	6.52	0.72
Cu-1	61.3 μm	53.74	0.15
Nb-1	30.0 μm	23.21	0.17
Al Degrader 01	4.96 mm	-	-
Al-2	25.5 μm	6.48	0.36
Cu-2	61.8 μm	53.85	0.17
Nb-2	30.8 μm	22.91	0.17
Al Degrader 02	4.55 mm	-	-
Al-3	25.8 μm	6.47	0.31
Cu-3	61.5 μm	53.98	0.11
Nb-3	31.0 μm	22.91	0.24
Al Degrader 03	3.52 mm	-	-
Al-4	26.3 μm	6.51	0.41
Cu-4	61.3 μm	53.46	0.22
Nb-4	30.8 μm	22.55	0.25
Al Degrader 04	3.47 mm	-	-
Al-5	26.5 μm	6.48	0.29
Cu-5	61.5 μm	53.57	0.11
Nb-5	30.8 μm	22.11	0.25
Al Degrader 05	3.46 mm	-	-
Al-6	26.3 μm	6.48	0.62
Cu-6	62.0 μm	53.84	0.32
Nb-6	31.3 μm	22.12	0.13
SS profile monitor	124.4 μm	101.34	0.23

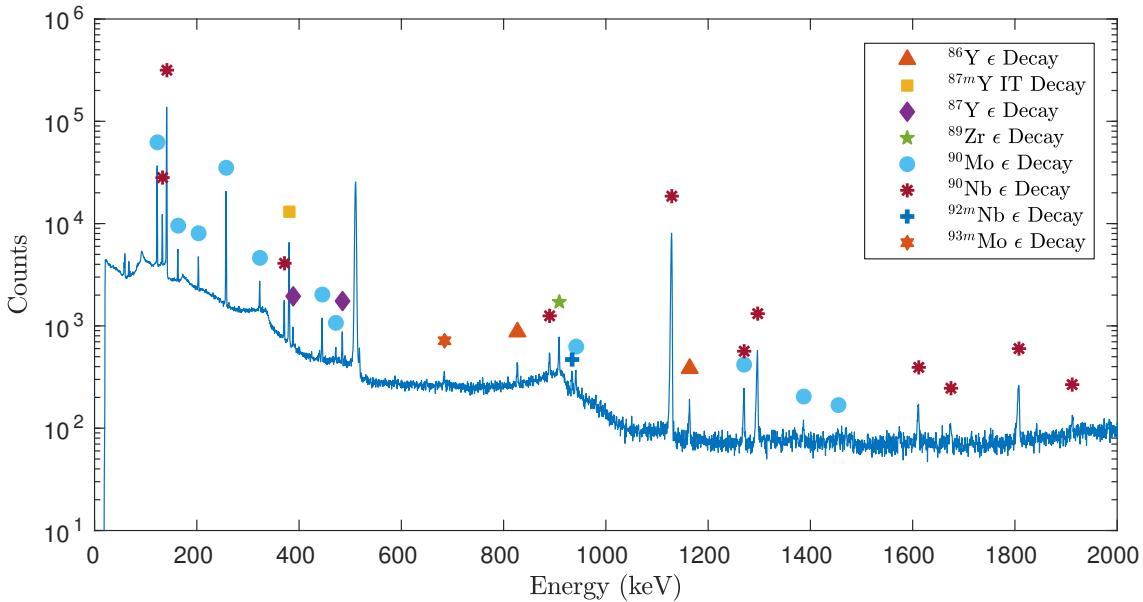


Figure 2.2: A gamma spectrum, collected from an activated Nb foil at approximately 80 MeV. While the majority of observed reaction products are visible in this spectrum, the ^{90}Mo decay lines, which form the basis of the $^{93}\text{Nb}(\text{p},\text{x})^{90}\text{Mo}$ monitor reaction, are high in intensity and clearly isolated from surrounding peaks.

Following acquisition, the decaying product nuclei corresponding to each observed peak in the collected spectra were identified. The calibrated detector efficiencies, along with gamma-ray intensities for each transition and corrections for gamma-ray attenuation within each foil packet, were used to convert the net counts in each fitted gamma-ray photopeak into an activity for the decay of the activation products. The nuclear decay data used in this work is tabulated in Table 2.6 and Table 2.7 of 2.7. Data for photon attenuation coefficients were taken from the XCOM photon cross sections database [31]. Decay gamma-rays from the product nuclei were measured at multiple points in time (up to 2 weeks after EoB), and as nearly all of the product nuclei have multiple high-intensity gamma-rays, this provided independent activity measurements at each time point. The total propagated uncertainty of the measured activity is the quadrature sum of the uncertainty in fitted peak areas, uncertainty in detector efficiency calibration, and uncertainty in the gamma-ray branching ratio data.

Since many of the reaction products populated by energetic protons are more than one decay off of stability, many of these are produced not only directly by reactions, but also indirectly by decay down a mass chain. To this end, it is useful to differentiate between the types of cross sections reported in this work. For the first observable product nucleus in a mass chain, its (p,x) cross section will be reported as a cumulative cross section (σ_c), which is the sum of direct production of that nucleus, as well as decay of its precursors and any other independent cross sections leading to that nucleus. Cumulative cross sections will be

reported whenever it is impossible to use decay spectrometry to distinguish independent production of a nucleus from decay feeding. For all remaining observed reaction products in the mass chain, and cases where no decay precursors exist, independent cross sections (σ_i) will be reported, allowing for determination of the independent production via subtraction and facilitating comparison to reaction model calculations.

Corrections must be made for the decay of the various reaction products during the time between EoB and the spectrum acquisition, in order to calculate A_0 , the initial activity at EoB, from the measured activities. The use of multiple gamma-rays at multiple points after EoB to calculate initial activities for each observed product nucleus allows for a more accurate determination of A_0 than simply basing its calculation off of a single gamma-ray observation. For the case of cumulative cross sections, EoB activities were quantified by fitting the activities observed at multiple time points t (since EoB) to the well-known radioactive decay law. Nonlinear regression was used for this fitting process, minimizing on χ^2 / degree of freedom, so that not only would the uncertainty-weighted EoB activities be fitted, but that a $1-\sigma$ confidence interval in A_0 could be reported as well. As with the gamma-ray intensities, all lifetimes used in this work are tabulated in Table 2.6 and Table 2.7 of 2.7. In the case of independent cross sections, a similar process was followed, quantifying $A_i(t=0) = A_{i,0}$, the EoB activity of nuclide i , by instead regressing to the solutions to the Bateman equation [32, 33]:

$$A_n(t) = \lambda_n \sum_{i=1}^n \left[N_{i,0} \times \left(\prod_{j=i}^{n-1} \lambda_j \right) \times \left(\sum_{j=i}^n \frac{e^{-\lambda_j t}}{\prod_{i \neq j} (\lambda_i - \lambda_j)} \right) \right] \quad (2.1)$$

where j refers to a precursor nucleus populating a specific end-product. While higher-order terms were added if needed, typically for an isomeric state in a particular mass chain, the second-order expansion ($n = 2$) was often sufficient to quantify EoB activities in a mass chain, simplifying to:

$$A_2(t) = \frac{A_{1,0}\lambda_2}{\lambda_1 - \lambda_2} (e^{-\lambda_2 t} - e^{-\lambda_1 t}) + A_{2,0}e^{-\lambda_2 t} \quad (2.2)$$

In these cases, the previously-quantified EoB activities from decay precursors ($A_{1,0}$, etc) would be substituted in, so that the feeding contributions from decay could be separated and an independent cross section reported. After quantifying the cumulative EoB activities at the top of a mass chain and all subsequent independent EoB activities, these will be later used to report the various cross sections for all observed reaction products and isomeric states.

Proton fluence determination

In addition to the LANSCE-IPF beamline's direct beam current measurements, thin ${}^{nat}\text{Al}$ and ${}^{nat}\text{Cu}$ foils were included along with the ${}^{nat}\text{Nb}$ targets at each energy position, to provide more sensitive beam current monitors. The IAEA-recommended ${}^{nat}\text{Al}(p,x){}^{22}\text{Na}$, ${}^{nat}\text{Al}(p,x){}^{24}\text{Na}$, ${}^{nat}\text{Cu}(p,x){}^{56}\text{Co}$, ${}^{nat}\text{Cu}(p,x){}^{62}\text{Zn}$, and ${}^{nat}\text{Cu}(p,x){}^{65}\text{Zn}$ monitor reactions were used for proton fluence measurement [11]. Due to the large energy degradation between the front and back

of the target stack, a non-trivial broadening of the proton energy distribution was expected for all monitor and target foils. As a result, the integral form of the well-known activation equation was used to accurately determine proton fluence ($I\Delta t$) in each monitor foil:

$$I\Delta t = \frac{A_0\Delta t}{\rho\Delta r (1 - e^{-\lambda\Delta t}) \int \sigma(E) \frac{d\phi}{dE} dE} \quad (2.3)$$

where A_0 is the EoB activity for the monitor reaction product, I is the proton current, $\rho\Delta r$ is the foil's areal density, λ is the monitor reaction product's decay constant, Δt is the length of irradiation, $\sigma(E)$ is the IAEA recommended cross section at energy E , and $\frac{d\phi}{dE}$ is the differential proton fluence. Using this formalism, the quantified EoB activities for each monitor reaction may be converted into a measured proton fluence at each energy position.

The propagated uncertainty in proton fluence is calculated as the quadrature sum of the uncertainty in quantified EoB activity, uncertainty in the duration of irradiation (conservatively estimated at 60 s, to account for any transient changes in beam current), uncertainty in foil areal density, uncertainty in monitor product half-life (included, but normally negligible), uncertainty in IAEA recommended cross section, and uncertainty in differential proton fluence. Of these, the first four contributions are all easily quantified in the preparation and execution of a stacked target irradiation; the last two contributions prove to be more nuanced, however. The uncertainty in proton fluence for irradiated monitor foils is derived from statistical uncertainty in the modeling of proton transport in the stack irradiation, discussed in section 2.4. The uncertainty in IAEA recommended cross section values must be estimated indirectly, as no uncertainty in the recommended cross sections is provided in the current IAEA evaluation. Fortunately, the recommended cross section values for each monitor reaction tend to closely match one of the selected experimental source data sets used in their evaluation. Since these data sets have listed uncertainties in the original manuscripts, uncertainties in IAEA recommended cross section values have been estimated by the uncertainty in the data set most closely matching the IAEA recommended values. For the monitor reactions employed in this work, these data sets are G. Steyn (1990) for ${}^{nat}\text{Al}(p,x){}^{22}\text{Na}$ [34], M. Uddin (2004) for ${}^{nat}\text{Al}(p,x){}^{24}\text{Na}$ [35], and S. Mills (1992) for ${}^{nat}\text{Cu}(p,x){}^{56}\text{Co}$, ${}^{nat}\text{Cu}(p,x){}^{62}\text{Zn}$, and ${}^{nat}\text{Cu}(p,x){}^{65}\text{Zn}$ [36].

Proton transport calculations

Initial estimates of the proton beam energy in all foils were calculated using the Anderson & Ziegler (A&Z) stopping power formalism [37–39]. These estimates of average beam energy in each foil are useful for the preliminary stack design. However, for final energy and fluence determinations, a more rigorous method of proton transport modeling is needed. The Monte Carlo N-Particle transport code MCNP6.1 was used for simulation of the full 3-D target stack, including determination of the full proton energy distribution for each stack position [40]. MCNP6 provides a far more robust method of proton transport, as it is able to account for beam losses due to scattering and reactions, as well as production of secondary particles.

As it is a Monte Carlo-based code, the uncertainty in energy distribution scales inversely with the number of source protons simulated. 10^8 source protons were used for all simulations, which places the statistical uncertainty in proton energy distributions at less than 0.01%.

The ability to model the full energy distribution in each target position is vital for stacked target irradiations, due to the progressively larger energy straggling towards the rear of the stack. The initial proton beam has a finite energy spread (an approximately 0.1 MeV Gaussian width at 100 MeV), and since stopping power for charged particles is inversely proportional to their energy, the low-energy tail of the energy distribution is degraded more in each stack element than the high-energy tail. This effect compounds towards the rear of the stack, creating a significantly broadened low-energy tail, and a progressively larger net shift of the centroid to a lower energy. To account for this increasing energy uncertainty, a suitably representative energy must be established for each foil in the target stack. In this work, the flux-weighted average proton energy in each foil, $\langle E \rangle$, represents the energy centroid for protons in a target stack component, calculated using the energy distributions $\frac{d\phi}{dE}$ from MCNP6 modeling of proton transport:

$$\langle E \rangle = \frac{\int E \frac{d\phi}{dE} dE}{\int \frac{d\phi}{dE} dE} \quad (2.4)$$

Likewise, to represent the energy uncertainty for each stack position, the full width at half maximum (FWHM) of the MCNP6-modeled energy distribution is chosen for each energy position reported. While most experimental uncertainties are reported at the 1σ level, the 2.355σ FWHM is used here to ensure at the 98% confidence interval that this width includes the “true” energy centroid value.

The “variance minimization” techniques described by Graves *et al.* have been employed here to further reduce the uncertainty in proton energy assignments [27]. This method is based on the assumption that the independent measurements of proton fluence from the five monitor reactions used in this work should all be consistent at each energy position. If the monitor reaction cross sections and MCNP6-modeled energy distributions are both accurate, disagreement in the observed proton fluences is due to poorly characterized stopping power in simulations, or a systematic error in the areal densities of the stack components [27, 41]. This disagreement is minor at the front of the stack, and gets progressively worse as the beam is degraded, due to the compounded effect of systematic uncertainties in stack areal densities.

Due to the significantly greater areal density of the thick 6061 aluminum degraders as compared to the other stack elements (nominal 3–5 mg/cm², relative to nominal 1000–1400 mg/cm²), the areal density of each of the 6061 aluminum degraders were varied uniformly in MCNP6 simulations by a factor of up to $\pm 25\%$ of nominal values, to find the effective density which minimized variance in the measured proton fluence at the lowest energy position (Al-6, Cu-6). This lowest energy position was chosen as a minimization candidate, as it is most sensitive to systematic uncertainties in stack design. The results of this minimization technique, shown in Figure 2.3, indicate a clear minimum in proton fluence variance for flux-

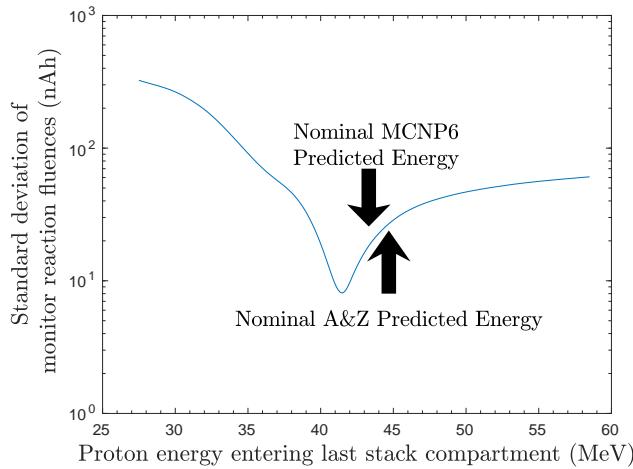


Figure 2.3: Result of the variance minimization performed by adjusting the degrader density in MCNP6 simulations of the target stack. A flux-weighted average proton energy of 41.34 MeV entering the last energy position creates a clear minimum in observed reaction fluence variance, corresponding to an areal density 2.52% greater than nominal. The variance minimum occurring at a lower incident energy than nominal MCNP6 and A&Z calculations indicates that there exists an additional systematic beam degradation not accounted for in modeling of proton transport in the stack design.

weighted average 41.34 MeV protons entering the last energy position. This is approximately 2 MeV lower than the nominal MCNP6 simulations, and approximately 3 MeV lower than nominal A&Z calculations, both of which used the nominal 2.80 g/cm^3 measured density of the 6061 aluminum degraders. This energy corresponds to a 6061 aluminum areal density of 2.52% greater than nominal measurements, and serves as a lump correction for other minor systematic uncertainties in stack design, including stack areal densities and incident beam energy.

The impact of this variance minimization is clearly seen in Figure 2.4. As expected, the 2.52% increase in 6061 aluminum areal density has an almost negligible impact on the higher-energy positions, but causes a progressively larger downshift in proton energies at the later energy positions. In addition, as one moves to the rear positions, the disagreement in the independent proton fluence measurements is reduced. It is worth noting that the proton fluence measured by the ${}^{nat}\text{Al}(p,x){}^{22}\text{Na}$ monitor reaction (threshold 21.0 MeV) is consistently higher in magnitude than all other monitor channels, with an increasing disparity at higher energies. This disparity is due to silicon in the Kapton tape (comprised of a silicone adhesive layer on a polyimide backing) used for sealing the foil packets, making up approximately 10% of the silicone on a stoichiometric basis. The ${}^{22}\text{Na}$ and ${}^{24}\text{Na}$ monitor channels can also be populated off of natural silicon (92.2% ${}^{28}\text{Si}$), predominantly via ${}^{28}\text{Si}(p,\alpha 2pn){}^{22}\text{Na}$ (threshold 35.3 MeV) and ${}^{28}\text{Si}(p,4pn){}^{24}\text{Na}$ (threshold 44.6 MeV). ${}^{29}\text{Si}$ and ${}^{30}\text{Si}$ are also potential targets for $(p,x){}^{22,24}\text{Na}$, albeit with higher energetic thresholds and smaller cross sections. The

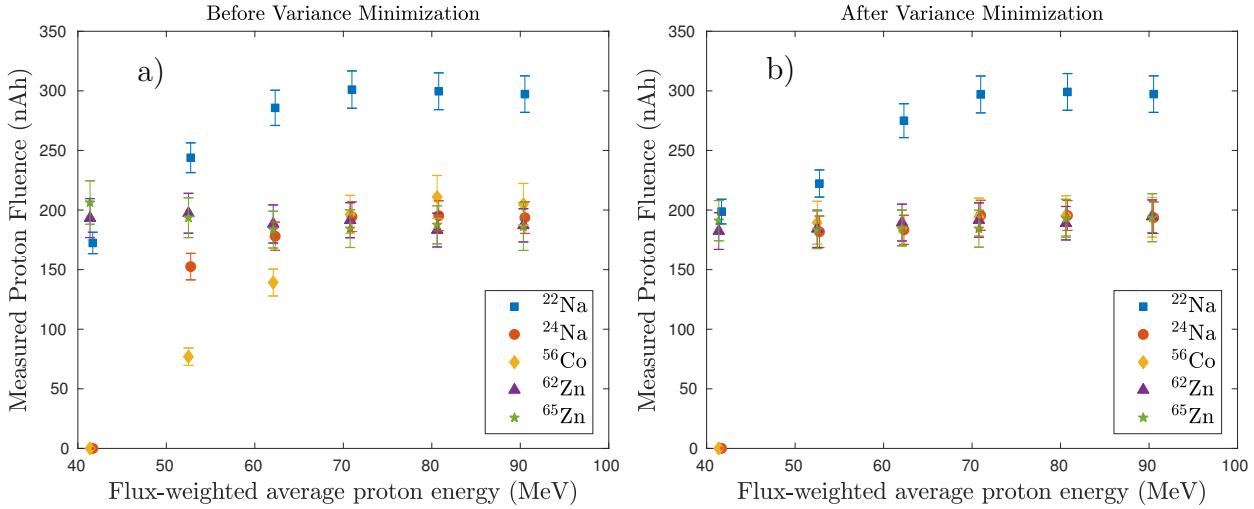


Figure 2.4: Results of variance minimization through enhancement of the effective areal density of the 6061 aluminum degraders by 2.52%. A noticeable reduction of variance in measured proton fluence is seen, particularly at the rear stack positions. Following minimization, additional apparent fluence is observed in the ${}^{nat}\text{Al}(p,x){}^{22}\text{Na}$ and ${}^{nat}\text{Al}(p,x){}^{24}\text{Na}$ monitor channels, due to contamination from ${}^{nat}\text{Si}(p,x){}^{22,24}\text{Na}$ on the silicone adhesive used for sealing foil packets.

attribution of excess $\text{Al}(p,x){}^{22,24}\text{Na}$ activity to the silicone adhesive is supported by the observation of ${}^{22}\text{Na}$ and ${}^{24}\text{Na}$ activities in all Cu and Nb foil positions.

${}^{nat}\text{Si}(p,\alpha 2\text{pn})$ is competitive with the ${}^{nat}\text{Al}(p,x)$ production route, seen when comparing the total measured activities of ${}^{22,24}\text{Na}$ in each Al foil packet, relative to the expected EoB activities for each reaction channel. This is presented in Figure 2.5. Since no evaluated cross section data exists in this energy region for ${}^{28}\text{Si}(p,x){}^{22}\text{Na}$ (and only minimal ${}^{nat}\text{Si}$ data exists), the TENDL-2015 library is used to estimate the expected relative EoB activities for ${}^{nat}\text{Al}(p,x){}^{22,24}\text{Na}$ and ${}^{nat}\text{Si}(p,x){}^{22,24}\text{Na}$, relative to IAEA recommended ${}^{nat}\text{Al}(p,x){}^{22,24}\text{Na}$ cross sections. Several observations are immediately obvious. At lower energies, the magnitude of ${}^{nat}\text{Al}(p,x){}^{22}\text{Na}$ is large compared to ${}^{nat}\text{Si}(p,x){}^{22}\text{Na}$, which is why the ${}^{nat}\text{Al}(p,x){}^{22}\text{Na}$ monitor agrees in fluence at the 40 (and almost at the 50) MeV position. At higher energies, the apparent ${}^{nat}\text{Al}(p,x){}^{22}\text{Na}$ activity begins to diverge from the IAEA expected activities as ${}^{nat}\text{Si}(p,x){}^{22}\text{Na}$ production begins to open up, which accounts for the nearly 50% apparent excess fluence in ${}^{22}\text{Na}$ between 60–90 MeV. For ${}^{24}\text{Na}$ production, we see similar behavior, with only a minor increase in apparent ${}^{24}\text{Na}$ activity, since the observed ${}^{nat}\text{Si}(p,x){}^{24}\text{Na}$ yield remains consistently low in magnitude. The observed ${}^{24}\text{Na}$ activities also follow the shape of the TENDL-2015 ${}^{nat}\text{Si}(p,x){}^{24}\text{Na}$ yields, albeit smaller in magnitude at the higher energy positions.

There are several important conclusions to be drawn from this simple estimate using the TENDL ${}^{nat}\text{Si}(p,x){}^{22,24}\text{Na}$ yields. The observation of the ${}^{22,24}\text{Na}$ activities in Cu and Nb

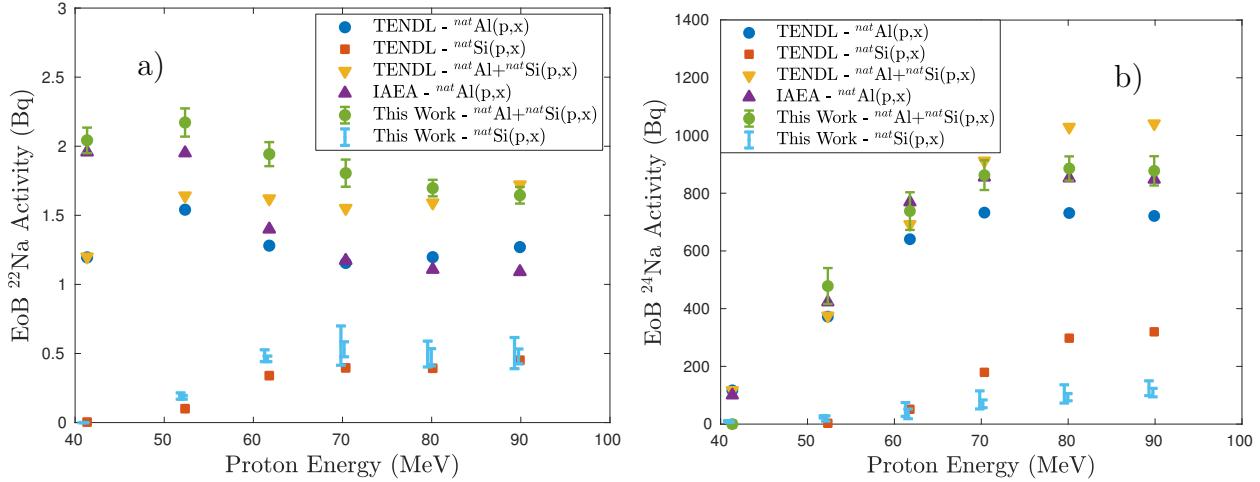


Figure 2.5: Estimates of EoB $^{nat}\text{Al}(p,x)^{22,24}\text{Na}$ and $^{nat}\text{Si}(p,x)^{22,24}\text{Na}$ activities using TENDL-2015 cross sections, in comparison with the IAEA recommended $^{nat}\text{Al}(p,x)^{22,24}\text{Na}$ cross sections. At low energies, experimentally observed apparent $^{22,24}\text{Na}$ activities in each Al foil packet are consistent with IAEA recommendations, but diverge at higher energies as the $^{nat}\text{Si}(p,x)^{22}\text{Na}$ exit channels begin to open up. $^{22,24}\text{Na}$ activities consistent with TENDL-2015 estimates are observed in each Nb and Cu foil packet as well, confirming that contamination may be attributed to activation of silicone adhesives.

foils represents an indirect measurement of the $^{nat}\text{Si}(p,x)^{22,24}\text{Na}$ cross sections, but will not be reported due to uncertainties in the areal density of the Si in the adhesive. However, if we assume a 10% Si stoichiometric basis and an areal density of 4.79 mg/cm^2 (based on bulk density), we can subtract out the measured $^{22,24}\text{Na}$ activity at each Nb and Cu foil position (correcting for the minor difference in proton energy between adjacent foils) from the apparent $^{22,24}\text{Na}$ activities observed in each Al foil packet, in order to obtain the “true” or uncontaminated fluence via the Al monitor reactions. The results of this may be seen in Figure 2.6. Following subtraction, the $^{22,24}\text{Na}$ fluences become more consistent with other monitor reaction channels, though ^{22}Na fluence remains 3–6% higher than the weighted mean of the remaining monitor reaction channels. While the dramatic improvement in monitor reaction consistency builds confidence, in the interest of surety and because they are consistent, only the $^{nat}\text{Cu}(p,x)^{56}\text{Co}$, $^{nat}\text{Cu}(p,x)^{62}\text{Zn}$, and $^{nat}\text{Cu}(p,x)^{65}\text{Zn}$ monitor reaction channels will be used for fluence determination for the reported cross sections. This serves as a pointed example of the importance of selecting monitor reaction products inaccessible through channels aside from the primary reaction ($^{nat}\text{Al}(p,x)^{22,24}\text{Na}$, in this case), as noted previously.

Using this variance minimized degrader density, the final incident proton energy energy distributions $\frac{d\phi}{dE}$ from MCNP6 simulation are shown for the six irradiated Nb foils in Figure 2.7. As expected, the energy distribution becomes increasingly more broadened at the lower energy

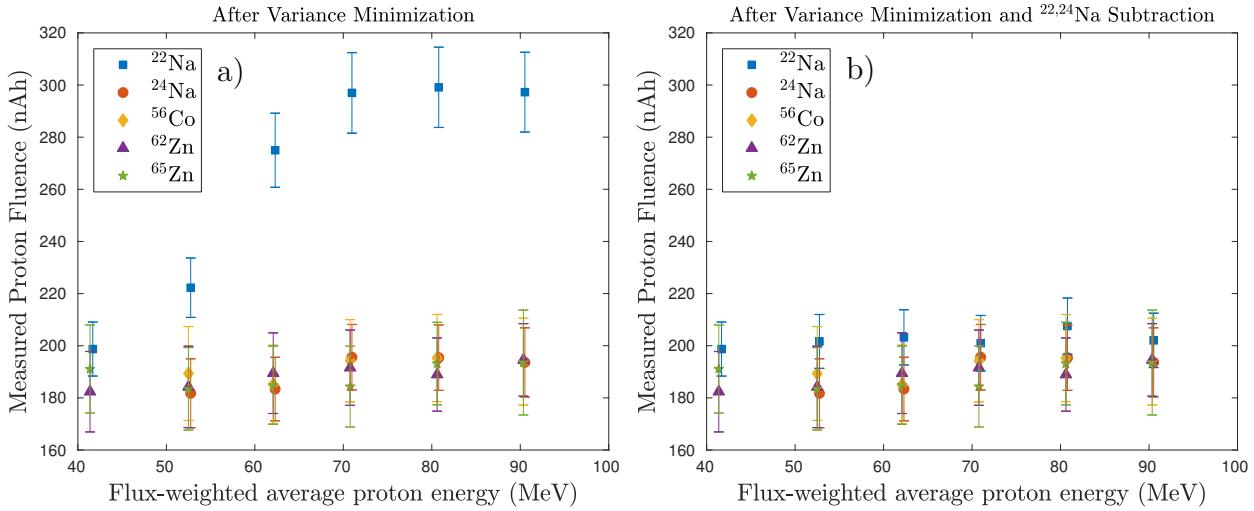


Figure 2.6: The “extra fluence” observed in the ${}^{\text{nat}}\text{Al}(\text{p},\text{x})^{22}\text{Na}$ and ${}^{\text{nat}}\text{Al}(\text{p},\text{x})^{24}\text{Na}$ monitor channels is caused by contamination from ${}^{\text{nat}}\text{Si}(\text{p},\text{x})^{22,24}\text{Na}$ on the silicone adhesive used for sealing foil packets. Following subtraction of ${}^{22,24}\text{Na}$ activities observed in the silicone adhesive of Nb and Cu foils in the same energy “compartment”, the consistency of the ${}^{\text{nat}}\text{Al}(\text{p},\text{x})^{22}\text{Na}$ monitor reaction improves dramatically. By excluding these contaminated channels, the remaining 3 independent monitor reactions serve to minimize uncertainty in stack energy assignments and incident fluence.

positions, as a result of the beam energy degradation. In addition, as the beam becomes more degraded, the magnitude of the peak of each energy distribution (as well as the integral of each distribution) is reduced, as beam fluence is lost due to scattering, and the peak-to-low-energy-tail ratio increases as more secondary protons are produced upstream. As with the monitor foils, these distributions were used to calculate the energy centroid (as the flux-weighted average proton energy) and uncertainty (as the FWHM of the distribution) for the final proton energy assignment of each Nb foil.

An enhanced version of the final ${}^{\text{nat}}\text{Cu}(\text{p},\text{x})^{56}\text{Co}$, ${}^{\text{nat}}\text{Cu}(\text{p},\text{x})^{62}\text{Zn}$, and ${}^{\text{nat}}\text{Cu}(\text{p},\text{x})^{65}\text{Zn}$ monitor reaction fluences may be seen in Figure 2.8. Without the reliable use of the ${}^{\text{nat}}\text{Al}(\text{p},\text{x})^{22}\text{Na}$ and ${}^{\text{nat}}\text{Al}(\text{p},\text{x})^{24}\text{Na}$ monitor channels, local interpolation cannot be used for fluence assignment to the Nb foils, and global interpolation is reliant upon a validated model for fluence loss. The uncertainty-weighted mean for the three ${}^{\text{nat}}\text{Cu}(\text{p},\text{x})$ monitor channels was calculated at each energy position, to determine the final fluence assignments for the Nb and Cu foils. Uncertainty in proton fluence is likewise calculated by error propagation of the fluence values at each energy position. These weighted-mean fluences are plotted in Figure 2.8, along with the estimated fluence according to both MCNP6 transport and an uncertainty-weighted linear χ^2 fit to the individual monitor channel fluence measurements. Both models reproduce the observed fluence data consistently within uncertainty, with the MCNP6 model predicting a slightly greater fluence loss throughout the stack. These models

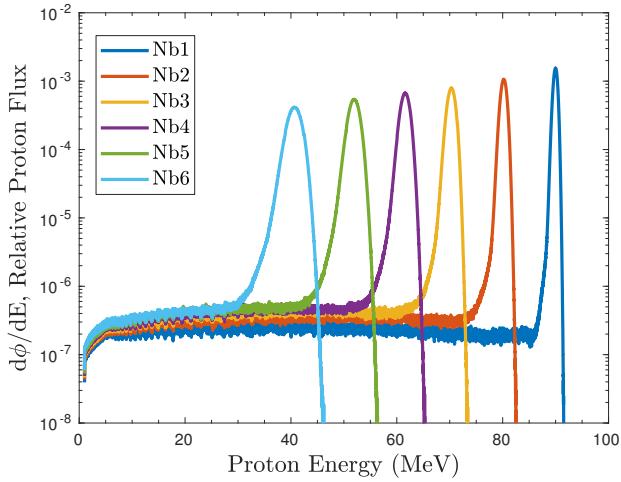


Figure 2.7: Final variance minimized incident proton energy distributions for the Nb foils, as simulated in MCNP6. The distribution tallies in each foil are all normalized to be per source proton, which was 10^8 in all simulations. As the beam is degraded, proton energy distributions become visibly broadened due to straggling, and drop in magnitude due to scattering losses.

are used purely to provide an extrapolation from the 90 MeV energy position back to the “front” of the stack at 100 MeV, to compare with the nominal fluence measured by IPF upstream current monitors.

Calculation of measured cross sections

Using the quantified EoB activities along with the variance-minimized proton fluence, it is possible to calculate the final cross sections for the various observed Nb(p,x) reactions. While thin (≈ 22 mg/cm²) Nb foils were irradiated to minimize the energy width of these cross section measurements, it is important to note that all cross sections reported here are flux-averaged over the energy distribution subtended by each foil, as seen in Figure 2.7. For both the cumulative and independent activities quantified, cross sections were calculated as:

$$\sigma = \frac{A_0}{\rho \Delta r I (1 - e^{-\lambda \Delta t})} \quad (2.5)$$

where A_0 is the EoB activity for the monitor reaction product, I is the proton current, $\rho \Delta r$ is the foil’s areal density, λ is the monitor reaction product’s decay constant, and Δt is the length of irradiation. The beam current, measured using an inductive pickup, remained stable for the duration of the irradiation, with the exception of approximately 70 s of downtime, occurring approximately 3 minutes into irradiation. The propagated uncertainty in cross section is calculated as the quadrature sum of the uncertainty in quantified EoB activity (which includes uncertainty in detector efficiencies), uncertainty in the duration of irradiation

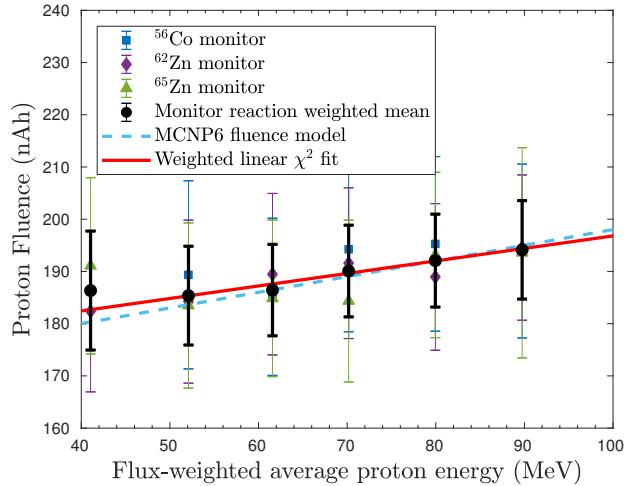


Figure 2.8: Final uncertainty-weighted mean proton fluences throughout the target stack, based on the variance-minimized observed fluence from the the ${}^{\text{nat}}\text{Cu}(p,x){}^{56}\text{Co}$, ${}^{\text{nat}}\text{Cu}(p,x){}^{62}\text{Zn}$, and ${}^{\text{nat}}\text{Cu}(p,x){}^{65}\text{Zn}$ monitor reactions. The fluence drops by approximately 7.2–8.9% from the incident fluence of $196.9\text{--}198.8$ nAh over the length of the target stack, based on fluence loss models from MCNP6 simulations and an empirical fit to fluence measurements.

(conservatively estimated at 60 s, to account for any transient changes in beam current), uncertainty in foil areal density, uncertainty in monitor product half-life (included, but normally negligible), and uncertainty in proton current (quantified by error propagation of the monitor reaction fluence values at each energy position, as seen in Figure 2.8).

2.5 Results

After irradiation, all foils were confirmed to still be sealed inside their Kapton packets, verifying that no activation products were lost due to packet failure and dispersal. In addition, each activated foil had a small “blister” under the Kapton tape layer, caused by a combination of thermal swelling and the formation of short-lived beta activities. This blister shows the location where the primary proton beam was incident upon the foil. The ${}^{\text{nat}}\text{Cu}(p,x){}^{56}\text{Co}$, ${}^{\text{nat}}\text{Cu}(p,x){}^{62}\text{Zn}$, and ${}^{\text{nat}}\text{Cu}(p,x){}^{65}\text{Zn}$ monitor reactions were used to determine the uncertainty-weighted mean fluence at each energy position (seen in Figure 2.8). A fluence of 198.8 ± 6.7 nAh was calculated to be incident upon the target stack using the MCNP6 fluence model, and a fluence of 196.9 ± 11.3 nAh using the linear fit model, both of which are consistent with the nominal fluence of 205.9 nAh based on IPF upstream current monitors. As fluence loss in the target box’s entrance window scales with $\sigma_{\text{tot}}\rho\Delta r$, it is expected that an extrapolation back to the stack entrance will underestimate the nominal fluence incident upon the box. This incident fluence dropped by approximately 8.9% to 180.9 ± 5.4 nAh (and by 7.2% to 182.7 ± 13.5 nAh using the linear fit model) over the length of the target stack,

Table 2.2: Measured cross sections for the various $^{nat}\text{Cu}(p,x)$ reaction products observed in this work. Cumulative cross sections are designated as σ_c , independent cross sections are designated as σ_i .

E_p (MeV)	Production cross section (mb)					
	$89.74^{+0.48}_{-0.43}$	$79.95^{+0.67}_{-0.64}$	$70.17^{+0.91}_{-0.85}$	$61.58^{+1.03}_{-0.98}$	$52.10^{+1.25}_{-1.20}$	$41.05^{+1.62}_{-1.54}$
^{51}Cr (σ_c)	0.919 ± 0.079	0.373 ± 0.023	0.450 ± 0.028	0.303 ± 0.016	—	—
^{52}Mn (σ_c)	1.70 ± 0.11	0.570 ± 0.031	0.0407 ± 0.0022	0.00526 ± 0.00057	—	—
^{52g}Mn (σ_i)	0.673 ± 0.043	0.239 ± 0.018	0.0164 ± 0.0023	0.000986 ± 0.000053	—	—
^{52m}Mn (σ_c)	1.023 ± 0.091	0.331 ± 0.030	0.0244 ± 0.0036	0.00427 ± 0.00052	—	—
^{54}Mn (σ_i)	5.87 ± 0.37	3.77 ± 0.21	4.14 ± 0.22	4.84 ± 0.26	1.680 ± 0.091	—
^{55}Co (σ_c)	1.71 ± 0.11	1.015 ± 0.058	0.193 ± 0.012	0.0299 ± 0.0028	0.00235 ± 0.00022	—
^{56}Ni (σ_c)	0.0806 ± 0.0051	0.1005 ± 0.0055	0.0906 ± 0.0046	0.0304 ± 0.0016	—	—
^{57}Ni (σ_c)	1.465 ± 0.093	1.202 ± 0.065	1.400 ± 0.071	2.13 ± 0.11	1.565 ± 0.083	0.0262 ± 0.0015
^{57}Co (σ_i)	40.1 ± 2.5	35.6 ± 1.9	35.8 ± 1.8	48.5 ± 2.5	47.7 ± 2.5	3.21 ± 0.18
^{58}Co (σ_c)	57.7 ± 4.5	55.0 ± 4.7	42.7 ± 3.4	33.7 ± 2.8	39.0 ± 3.8	62.3 ± 4.6
^{58g}Co (σ_i)	14.0 ± 2.5	10.8 ± 2.1	6.1 ± 1.6	7.8 ± 1.4	7.1 ± 1.7	1.12 ± 0.32
^{58m}Co (σ_i)	43.6 ± 3.7	44.2 ± 4.3	36.6 ± 3.0	25.8 ± 2.5	31.9 ± 3.3	61.1 ± 4.6
^{59}Fe (σ_c)	0.865 ± 0.057	0.837 ± 0.046	0.749 ± 0.039	0.616 ± 0.034	0.209 ± 0.014	—
^{60}Co (σ_c)	13.23 ± 0.87	13.47 ± 0.78	11.14 ± 0.94	11.44 ± 0.80	9.30 ± 0.87	6.6 ± 1.1
^{61}Cu (σ_c)	50.5 ± 3.3	56.1 ± 3.2	65.1 ± 3.6	72.2 ± 4.0	80.6 ± 4.7	157.1 ± 8.6
^{64}Cu (σ_i)	38.7 ± 2.7	42.8 ± 2.4	45.5 ± 2.7	50.2 ± 2.8	55.7 ± 3.0	63.3 ± 3.6

which is consistent with similar measurements at IPF in the past [27]. This loss of fluence is due to a combination of (p,x) reactions throughout the target stack, as well as large-angle deflections (primarily in the aluminum degraders) from scattering of the beam.

Using the final proton fluence at each energy position, cross sections for ^{51}Cr , ^{52g}Mn , ^{52m}Mn , ^{54}Mn , ^{55}Co , ^{56}Ni , ^{57}Ni , ^{57}Co , ^{58g}Co , ^{58m}Co , ^{59}Fe , ^{60}Co , ^{61}Cu , and ^{64}Cu were extracted for (p,x) reactions on ^{nat}Cu foils in the 40–90 MeV region, as recorded in Table 2.2. For (p,x) reactions on ^{nat}Nb foils, the (p,x) cross sections for ^{82m}Rb , ^{83}Sr , ^{85g}Y , ^{85m}Y , ^{86}Zr , ^{86}Y , ^{87}Zr , ^{87g}Y , ^{87m}Y , ^{88}Zr , ^{88}Y , ^{89g}Nb , ^{89m}Nb , ^{89}Zr , ^{90}Mo , ^{90}Nb , ^{91m}Nb , ^{92m}Nb , and ^{93m}Mo were extracted, as recorded in Table 2.3. In addition, as there exist a number of isomers with radioactive ground states in these mass regions, independent measurements of isomer-to-ground-state branching ratios for $^{52m/g}\text{Mn}$, $^{58m/g}\text{Co}$, $^{85m/g}\text{Y}$, $^{87m/g}\text{Y}$, and $^{89m/g}\text{Nb}$ were extracted and are recorded in Table 2.4. Comparisons of the measured cross sections and isomer branching ratios with literature data (retrieved from EXFOR [42]) are seen in the figures of 2.8 and 2.9. The propagated uncertainty in these cross sections varies widely based on the reaction product in question, with the major components arising from uncertainty in EoB activity ($\pm 3\text{--}7\%$), proton fluence ($\pm 4\text{--}6\%$), and foil areal density ($\pm 0.1\text{--}0.6\%$).

These results have several notable features. The various $^{nat}\text{Cu}(p,x)$ cross sections measured here are in excellent agreement with the body of measurements in the literature, but have been measured nearly exclusively with the highest precision to date. Similarly, the various $^{nat}\text{Nb}(p,x)$ cross sections measured here are in excellent agreement with literature data, which is far more sparse in the 40–90 MeV region than for $^{nat}\text{Cu}(p,x)$ — fewer than three existing measurements have been performed for the majority of the reactions presented here. Indeed,

Table 2.3: Measured cross sections for the various $^{nat}\text{Nb}(p,x)$ reaction products observed in this work. Cumulative cross sections are designated as σ_c , independent cross sections are designated as σ_i .

E_p (MeV)	Production cross section (mb)					
	$89.37_{-0.45}^{+0.47}$	$79.55_{-0.64}^{+0.68}$	$69.70_{-0.85}^{+0.90}$	$61.07_{-0.98}^{+1.05}$	$51.51_{-1.21}^{+1.25}$	$40.34_{-1.55}^{+1.58}$
^{82m}Rb (σ_c)	2.48 ± 0.22	—	—	—	—	—
^{83}Sr (σ_c)	4.02 ± 0.61	4.78 ± 0.42	3.49 ± 0.36	—	—	—
^{85}Y (σ_c)	13.78 ± 0.55	7.52 ± 0.51	2.11 ± 0.14	—	—	—
^{85g}Y (σ_i)	2.37 ± 0.11	2.08 ± 0.17	0.557 ± 0.037	—	—	—
^{85m}Y (σ_i)	11.41 ± 0.54	5.44 ± 0.48	1.55 ± 0.13	—	—	—
^{86}Zr (σ_c)	12.68 ± 0.68	18.21 ± 0.93	19.28 ± 0.97	6.16 ± 0.32	—	—
^{86}Y (σ_i)	33.4 ± 1.8	41.6 ± 2.2	39.9 ± 2.1	13.56 ± 0.72	—	—
^{87}Zr (σ_c)	47.4 ± 7.3	28.0 ± 2.8	32.2 ± 2.9	49.8 ± 5.0	38.2 ± 3.7	1.12 ± 0.17
^{87}Y (σ_i)	110.0 ± 7.2	54.7 ± 2.8	61.0 ± 2.9	90.0 ± 4.9	67.2 ± 3.6	2.91 ± 0.17
^{87g}Y (σ_i)	28.0 ± 5.8	7.4 ± 1.3	6.55 ± 0.64	5.8 ± 2.2	2.63 ± 0.47	0.942 ± 0.073
^{87m}Y (σ_i)	82.0 ± 4.3	47.3 ± 2.5	54.4 ± 2.8	84.2 ± 4.4	64.6 ± 3.6	1.97 ± 0.15
^{88}Zr (σ_c)	159.1 ± 7.8	144.6 ± 6.8	62.4 ± 3.1	21.2 ± 1.0	33.6 ± 1.8	65.3 ± 4.0
^{88}Y (σ_i)	17.2 ± 1.1	13.27 ± 0.86	7.98 ± 0.72	2.91 ± 0.25	9.2 ± 1.4	9.88 ± 0.69
^{89}Nb (σ_c)	—	—	179 ± 14	214.4 ± 9.8	—	—
^{89g}Nb (σ_i)	—	—	145 ± 14	186.4 ± 9.6	—	—
^{89m}Nb (σ_i)	—	—	34.7 ± 2.6	28.0 ± 2.0	—	—
^{89}Zr (σ_i)	211 ± 11	243 ± 13	294 ± 15	257 ± 13	55.4 ± 3.0	15.5 ± 1.0
^{90}Mo (σ_i)	21.3 ± 1.1	26.4 ± 1.3	34.5 ± 1.6	61.9 ± 3.1	122.0 ± 6.1	24.2 ± 1.5
^{90}Nb (σ_i)	158.3 ± 8.1	174.9 ± 8.5	209.3 ± 9.9	272 ± 14	369 ± 19	163.9 ± 9.8
^{91m}Nb (σ_c)	—	—	—	—	—	66.5 ± 5.8
^{92m}Nb (σ_i)	43.7 ± 2.4	47.3 ± 2.4	49.8 ± 2.6	52.9 ± 2.8	55.3 ± 3.1	59.9 ± 3.9
^{93m}Mo (σ_i)	0.97 ± 0.20	1.29 ± 0.15	1.62 ± 0.24	1.85 ± 0.15	1.86 ± 0.14	2.00 ± 0.15

 Table 2.4: Measured isomer-to-ground-state branching ratios for the various $^{nat}\text{Nb}(p,x)$ and $^{nat}\text{Cu}(p,x)$ reaction products observed in this work.

E_p (MeV)	Isomer branching ratio					
	$89.74_{-0.43}^{+0.48}$	$79.95_{-0.64}^{+0.67}$	$70.17_{-0.85}^{+0.91}$	$61.58_{-0.98}^{+1.03}$	$52.10_{-1.20}^{+1.25}$	$41.05_{-1.54}^{+1.62}$
$^{nat}\text{Cu}(p,x)^{52}\text{Mn}$	0.603 ± 0.066	0.581 ± 0.062	0.598 ± 0.095	0.81 ± 0.13	—	—
$^{nat}\text{Cu}(p,x)^{58}\text{Co}$	0.757 ± 0.088	0.80 ± 0.10	0.858 ± 0.099	0.767 ± 0.097	0.82 ± 0.12	0.98 ± 0.10
E_p (MeV)	$89.37_{-0.45}^{+0.47}$	$79.55_{-0.64}^{+0.68}$	$69.70_{-0.85}^{+0.90}$	$61.07_{-0.98}^{+1.05}$	$51.51_{-1.21}^{+1.25}$	$40.34_{-1.55}^{+1.58}$
$^{nat}\text{Nb}(p,x)^{85}\text{Y}$	0.828 ± 0.051	0.724 ± 0.080	0.736 ± 0.080	—	—	—
$^{nat}\text{Nb}(p,x)^{87}\text{Y}$	0.746 ± 0.063	0.865 ± 0.063	0.893 ± 0.063	0.936 ± 0.070	0.961 ± 0.075	0.676 ± 0.065
$^{nat}\text{Nb}(p,x)^{89}\text{Nb}$	—	—	0.193 ± 0.021	0.130 ± 0.011	—	—

the ${}^{\text{nat}}\text{Nb}(\text{p},\text{x}) {}^{83}\text{Sr}$, ${}^{\text{nat}}\text{Nb}(\text{p},\text{x}) {}^{85}\text{Y}$, ${}^{\text{nat}}\text{Nb}(\text{p},\text{x}) {}^{89}\text{Nb}$, ${}^{\text{nat}}\text{Nb}(\text{p},\text{x}) {}^{90}\text{Mo}$, ${}^{\text{nat}}\text{Nb}(\text{p},\text{x}) {}^{91\text{m}}\text{Nb}$, and ${}^{\text{nat}}\text{Nb}(\text{p},\text{x}) {}^{98\text{m}}\text{Mo}$ reactions each possess no more than a total of three data points in this energy region. Not only do the ${}^{\text{nat}}\text{Nb}(\text{p},\text{x})$ measurements in this work fill in the sparse data in this energy region, but they have been measured with the highest precision relative to existing literature data.

This work presents the first measurements of several observables in this mass region, including the ${}^{\text{nat}}\text{Nb}(\text{p},\text{x}) {}^{82\text{m}}\text{Rb}$ reaction in the 40–90 MeV region, the independent cross section for ${}^{\text{nat}}\text{Cu}(\text{p},\text{x}) {}^{52\text{g}}\text{Mn}$, and the ${}^{52\text{m}}\text{Mn}$ (2^+) / ${}^{52\text{g}}\text{Mn}$ (6^+) isomer branching ratio via ${}^{\text{nat}}\text{Cu}(\text{p},\text{x})$. The cumulative cross sections from these data are also consistent with existing measurements of the cumulative ${}^{\text{nat}}\text{Cu}(\text{p},\text{x}) {}^{52}\text{Mn}$ cross section. Similarly, this work offers the first measurement of the independent cross sections for ${}^{\text{nat}}\text{Nb}(\text{p},\text{x}) {}^{85\text{g}}\text{Y}$, as well as the first measurement of the ${}^{85\text{m}}\text{Y}$ ($9/2^+$) / ${}^{85\text{g}}\text{Y}$ ($1/2^-$) isomer branching ratio via ${}^{\text{nat}}\text{Nb}(\text{p},\text{x})$.

Notably, this work is the most well-characterized measurement of the ${}^{\text{nat}}\text{Nb}(\text{p},\text{x}) {}^{90}\text{Mo}$ reaction below 100 MeV to date, with cross sections measured at the 4–6% uncertainty level. This is important, as it presents the first step towards characterizing this reaction for use as a proton monitor reaction standard below 100 MeV. ${}^{\text{nat}}\text{Nb}(\text{p},\text{x}) {}^{90}\text{Mo}$ can only be populated through the (p,4n) reaction channel, so no corrections for (n,x) contamination channels or decay down the A=90 isobar are needed. ${}^{90}\text{Mo}$ possesses seven strong, distinct gamma lines which can easily be used for its identification and quantification. Finally, the production of ${}^{90}\text{Mo}$ in the 40–90 MeV region is quite strong, with a peak cross section of approximately 120 mb. Combining the reaction yield and gamma abundance, the use of approximately 23 mg/cm² Nb targets easily provided sufficient counting statistics for activity quantification in the 40–90 MeV region. This result presents the first step towards the use of ${}^{90}\text{Mo}$ as a clean and precise charged particle monitor reaction standard in irradiations up to approximately 24 hours in duration.

In addition to the ${}^{\text{nat}}\text{Nb}(\text{p},\text{x}) {}^{90}\text{Mo}$ measurement, this experiment has also yielded measurements of a number of additional emerging radionuclides with medical applications. These include the non-standard positron emitters ${}^{57}\text{Ni}$ [27, 43–45], ${}^{64}\text{Cu}$ [46–53], ${}^{86}\text{Y}$ [19, 26, 54–62], ${}^{89}\text{Zr}$ [63–67], ${}^{90}\text{Nb}$ [68, 69], and the Auger-therapy agent ${}^{82\text{m}}\text{Rb}$ [70, 71]. Production of these radionuclides offers no major advantages over established pathways, with the generally lower yields and radioisotopic purities failing to justify the convenience of natural targets via ${}^{\text{nat}}\text{Cu}(\text{p},\text{x})$ and ${}^{\text{nat}}\text{Nb}(\text{p},\text{x})$. The one possible exception to this trend is the non-standard positron emitter ${}^{57}\text{Ni}$ ($t_{1/2} = 35.60 \pm 0.06$ h, $\epsilon=100\%$ to ${}^{57}\text{Co}$ [16]) — the ${}^{57}\text{Ni}/{}^{56}\text{Ni}$ ratio of production rates is approximately 290 at 61.58 MeV, and varies from 45–75 at the 70–90 MeV positions. This ${}^{\text{nat}}\text{Cu}(\text{p},\text{x})$ route offers both higher yield and higher radioisotopic purity over the established ${}^{\text{nat}}\text{Co}(\text{p},3\text{n})$ pathway, which suffers from approximately five-fold greater ${}^{56}\text{Ni}$ contamination [72, 73].

We wish to urge caution in future stacked-target activation experiments by avoiding the use of silicone adhesive-based tapes for foil containment, especially when paired with the use of Al monitor foils. Acrylic-based tape options are commercially available, and are immune from (p,x) production of ${}^{22,24}\text{Na}$ activities, due to being of too low-Z for these reaction channels to be possible. Even with subtraction of ${}^{22,24}\text{Na}$ activities though irradiating a

Table 2.5: Default settings for the reactions codes

<u>Code Version</u>	<u>Proton/Neutron Optical Model</u>	<u>Alpha Optical Model</u>	<u>E1 γSF Model</u>
EMPIRE-3.2.3[75]	Koning-Delaroche[76]	Avrigeanu(2009)[77]	Modified Lorentzian[78]
TALYS-1.8[79]	Koning-Delaroche	Specific folded potential[79]	Brink-Axel Lorentzian[79]
CoH-3.5.1[80, 81]	Koning-Delaroche	Avrigeanu(1994)[82]	Generalized Lorentzian[80, 81]

Kapton tape “blank” or similar, we observe the Al monitor channels to measure consistently higher proton fluence than via Cu monitor channels, by 5–8%. If Al monitors are used in conjunction with silicone-based tapes, even with subtraction of excess ^{22}Na activities, a systematically enhanced fluence may be determined, leading to cross sections reported with inaccurately diminished magnitude. Furthermore, since data for monitor reactions are often self-referencing, the propagated impact of this systematic enhancement in fluence may have far-reaching consequences for both medical isotope production, as well as for the evaluated nuclear data libraries, which use these proton activation experiments as input.

As mentioned before, cumulative cross sections are reported here for the first observable product nuclei in a mass chain, or whenever it is impossible to use decay spectrometry to distinguish direct production of a nucleus from decay feeding. For all remaining observed reaction products in the mass chain, and cases where no decay precursors exist, independent cross sections are reported, allowing for determination of the direct production via subtraction. This, in turn, offers the opportunity to gauge the predictive capabilities of modern nuclear models used in the reaction evaluation process. The reaction channels with independent cross sections were compared to calculations with the reaction modeling codes EMPIRE, TALYS, and CoH, run with the default settings. The default optical models and E1 gamma strength function models for each code are presented in Table 2.5. The large energy range covered by many of the exit channels, which extends significantly beyond the range of pure compound nuclear/evaporation, allows the data to be used to study the differences between these modeling codes in the pre-equilibrium regime.

The default level density in both CoH and TALYS is the Gilbert-Cameron model, which uses a Constant Temperature model below a critical energy and Fermi Gas model above it. The default level density in EMPIRE is the Enhanced Generalized Superfluid Model (EGSM) which uses the Generalized Superfluid model below a critical energy, and Fermi Gas model above it [74]. The EGSM densities are normalized to D_0 and the discrete levels, but in such a way that only the level density below the neutron separation energy is effected by the discrete levels chosen for the normalization. All three codes use a two-exciton phenomenological model to calculate the pre-equilibrium cross section, but the specific implementation differs between the codes.

Given the large number of exit channels in this data set, we will limit our discussion to cross sections for the production of a specific residual nucleus with experimental data through the full rise and fall of the peak, and at least 1% of the total reaction cross section. Exit channel cross sections that do not exhibit the full rise and fall of the peak, which is identified as being dominated by the formation of a compound nucleus, do not provide enough

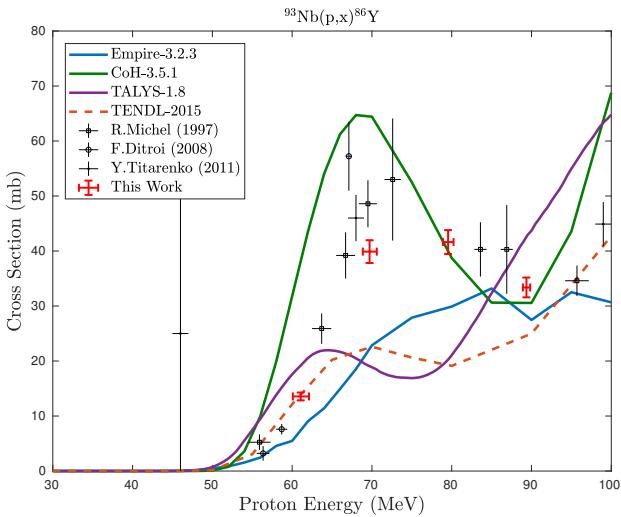


Figure 2.9: Measured $^{93}\text{Nb}(p,x)^{86}\text{Y}$ cross section, with the $^{93}\text{Nb}(p,\alpha p3n)^{86}\text{Y}$ reaction channel visibly peaking at approximately 70 MeV.

information to analyze the calculations. Residual nuclei like ^{88}Zr that can be produced by multiple reaction channels, such as $(p,\alpha 2n)$ and by $(p,2p4n)$ are also not discussed in depth. We exclude reactions with cross sections with peak values less than 1% of the total reaction cross section because their behavior is extremely sensitive to more dominant channels. The three residual nuclei that meet all of the above criteria for which there is an independent measurement of the residual production cross section are ^{86}Y , ^{90}Mo , and ^{90}Nb .

The $^{93}\text{Nb}(p,\alpha p3n)^{86}\text{Y}$ reaction channel, which peaks at approximately 70 MeV, is well within the compound regime for the entire energy region of this experiment. The data collected on this residual is consistent with the one other data set available, taken in 1997 by Michel *et al.* [72]. The $^{93}\text{Nb}(p,4n)^{90}\text{Mo}$ and $^{93}\text{Nb}(p,p3n)^{90}\text{Nb}$ channels both peak early in the energy region, around 50 MeV, and the data clearly show the full rise, peak, and fall of the compound cross section. In both of these channels, this data is consistent with the data by Titarenko *et al.* in 2011[71].

In all three channels, the TALYS, TENDL, and CoH calculations rise, peak, and fall at lower energies than the data, while EMPIRE calculates the peak to occur at higher energies. For ^{90}Mo , the EMPIRE peak is representative of the data. For ^{86}Y and ^{90}Nb , the peak is missed by all three of the codes.

The magnitudes of the TALYS and TENDL calculations are consistently too low in the three channels studied here. For ^{86}Y , CoH and EMPIRE also predict smaller cross sections than the data would suggest, which may be influenced by incorrect modeling of other, stronger, channels. The magnitude of the peak in the CoH calculation for ^{90}Mo is consistent with the data, while EMPIRE predicts a cross section that is approximately the same magnitude as that of TALYS. ^{90}Nb is one of the strongest measured channels, approximately 10% of the total reaction cross section, and the values from the three codes are all consistent, but too

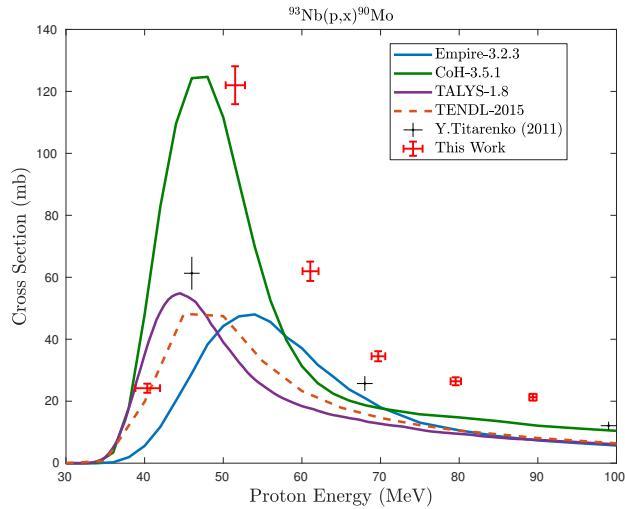


Figure 2.10: Measured $^{93}\text{Nb}(p,x)^{90}\text{Mo}$ cross section, with the $^{93}\text{Nb}(p,4n)^{90}\text{Mo}$ reaction channel visibly peaking at approximately 50 MeV.

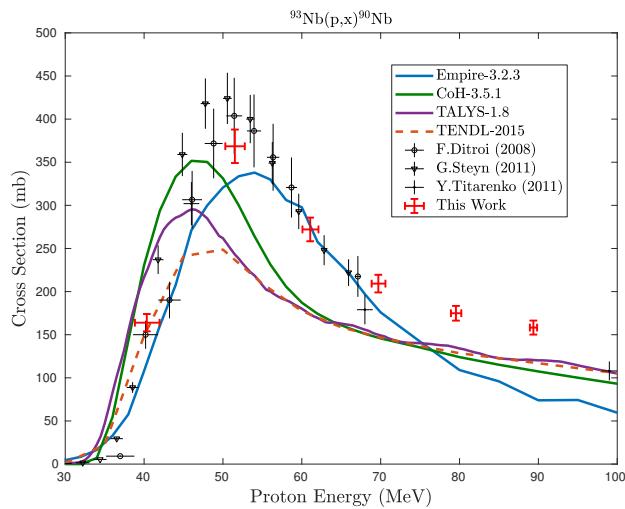


Figure 2.11: Measured $^{93}\text{Nb}(p,x)^{90}\text{Nb}$ cross section, with the $^{93}\text{Nb}(p,p3n)^{90}\text{Nb}$ reaction channel visibly peaking at approximately 50 MeV.

small, in magnitude.

The ^{90}Nb production cross section exhibits a persistent pre-equilibrium “tail” that keeps the channel open well after the compound cross section has fallen away. TALYS, TENDL, and CoH seem to have the correct shape for this pre-equilibrium cross section, with magnitudes that are just slightly too low. EMPIRE, however, does not level off as much as the data and the other codes are seen to, and misses the high-energy data points.

2.6 Conclusions

We present here a set of 38 measurements of cross sections for the $^{\text{nat}}\text{Nb}(p,x)$ and $^{\text{nat}}\text{Cu}(p,x)$ reactions between 40–90 MeV, as well as five independent measurements of isomer branching ratios. Nearly all cross sections have been reported with higher precision than previous measurements. We report the first measurements of the $^{\text{nat}}\text{Nb}(p,x)^{82\text{m}}\text{Rb}$ reaction, as well as the first measurement of the independent cross sections for $^{\text{nat}}\text{Cu}(p,x)^{52\text{m}}\text{Mn}$, $^{\text{nat}}\text{Cu}(p,x)^{52\text{g}}\text{Mn}$, and $^{\text{nat}}\text{Nb}(p,x)^{85\text{g}}\text{Y}$ in the 40–90 MeV region. We advise that future activation experiments avoid the use of silicone-based adhesives, particularly in conjunction with aluminum monitor foils, to avoid reporting an enhanced fluence due to $^{22,24}\text{Na}$ contamination. We also use these measurements to illustrate the deficiencies in the current state of reaction modeling for 40–90 MeV $^{\text{nat}}\text{Nb}(p,x)$ and $^{\text{nat}}\text{Cu}(p,x)$ reactions. Finally, this work provides another example of the usefulness of the recently-described variance minimization techniques for reducing energy uncertainties in stacked target charged particle irradiation experiments.

2.7 Decay data

The lifetimes and gamma-ray branching ratios listed in these tables were used for all calculations of measured cross sections reported in this work, and have been taken from the most recent edition of Nuclear Data Sheets for each mass chain [15–20, 23–26, 83–98].

Table 2.6: Decay data for gamma-rays observed in ${}^{\text{nat}}\text{Al(p,x)}$ and ${}^{\text{nat}}\text{Cu(p,x)}$.

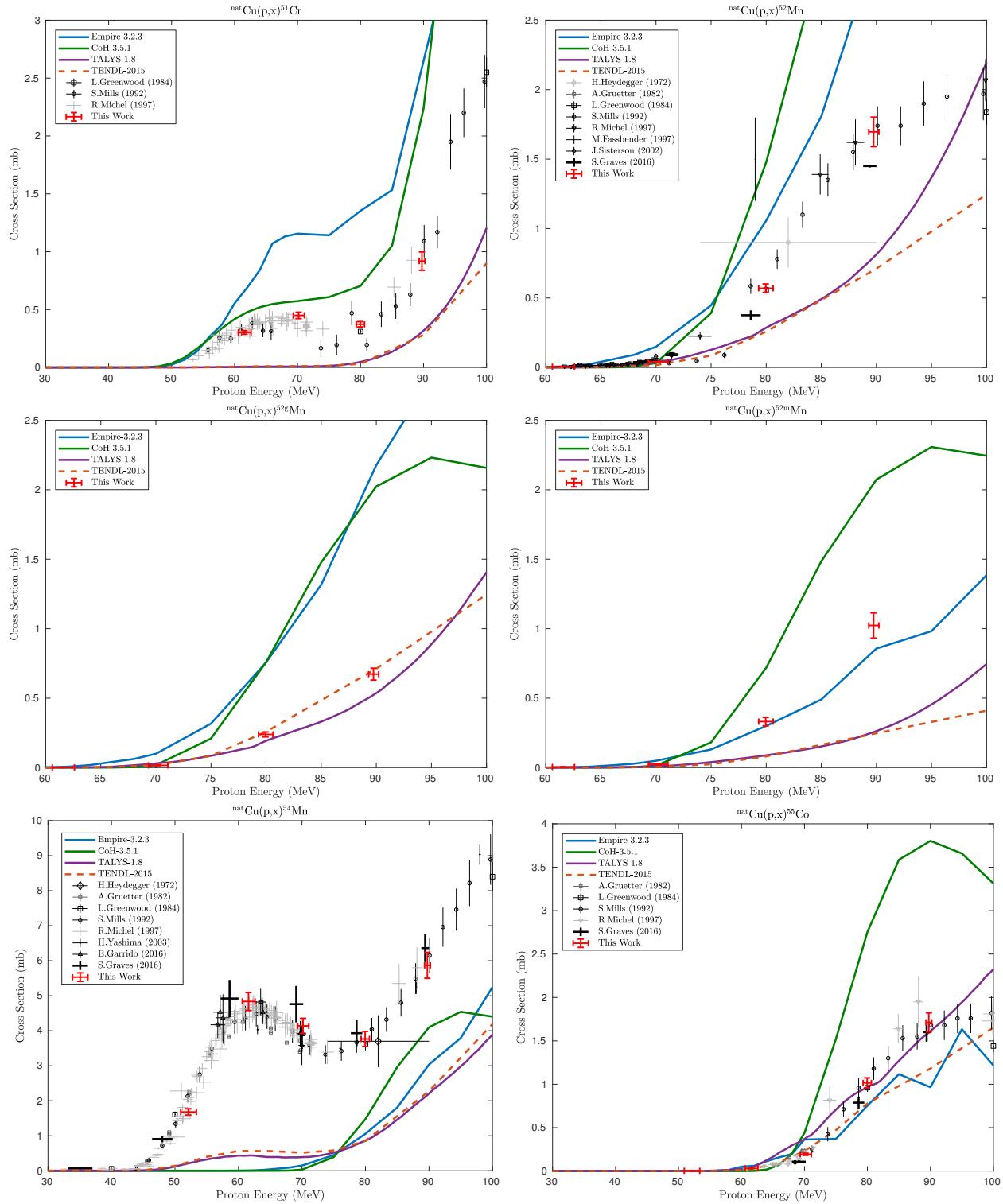
Nuclide	Half-life	E_γ (keV)	I_γ (%)
${}^{22}\text{Na}$	2.6018(22) y	1274.537	99.940(14)
${}^{24}\text{Na}$	14.997(12) h	1368.626	99.9936(15)
${}^{51}\text{Cr}$	27.704(3) d	320.0824	9.910(10)
${}^{52m}\text{Mn}$	21.1(2) m	1434.0600	98.2(5)
${}^{52}\text{Mn}$	5.591(3) d	744.233	90.0(12)
	5.591(3) d	935.544	94.5(13)
	5.591(3) d	1246.278	4.21(7)
	5.591(3) d	1434.092	100.0(14)
${}^{54}\text{Mn}$	312.20(20)	834.848	99.9760(10)
${}^{55}\text{Co}$	17.53(3) h	477.2	20.2(17)
	17.53(3) h	931.1	75.0(35)
	17.53(3) h	1316.6	7.1(3)
	17.53(3) h	1408.5	16.9(8)
${}^{56}\text{Ni}$	6.075(10) d	158.38	98.8(10)
	6.075(10) d	269.50	36.5(8)
	6.075(10) d	480.44	36.5(8)
	6.075(10) d	749.95	49.5(12)
	6.075(10) d	811.85	86.0(9)
	6.075(10) d	1561.80	14.0(6)
${}^{56}\text{Co}$	77.236(26) d	846.770	99.9399(2)
	77.236(26) d	1037.843	14.05(4)
	77.236(26) d	1238.288	66.46(12)
	77.236(26) d	1360.212	4.283(12)
	77.236(26) d	1771.357	15.41(6)
${}^{57}\text{Ni}$	35.60(6) h	127.164	16.7(5)
	35.60(6) h	1377.63	81.7(24)
	35.60(6) h	1757.55	5.75(20)
	35.60(6) h	1919.52	12.3(4)
${}^{57}\text{Co}$	271.74(6) d	122.06065	85.60(17)
	271.74(6) d	136.47356	10.68(8)
${}^{58}\text{Co}$	70.86(6) d	810.7593	99.450(10)
	70.86(6) d	863.951	0.686(10)
${}^{59}\text{Fe}$	44.495(9) d	1099.245	56.5(18)
	44.495(9) d	1291.590	43.2(14)
${}^{60}\text{Co}$	5.2714(5) y	1173.228	99.85(3)
	5.2714(5) y	1332.492	99.9826(6)
${}^{61}\text{Cu}$	3.339(8) h	282.956	12.2(2.2)
	3.339(8) h	373.050	2.1(4)
	3.339(8) h	656.008	10.8(20)
	3.339(8) h	1185.234	3.7(7)
${}^{62}\text{Zn}$	9.193(15) h	243.36	2.52(23)
	9.193(15) h	246.95	1.90(18)
	9.193(15) h	260.43	1.35(13)
	9.193(15) h	394.03	2.24(17)
	9.193(15) h	548.35	15.3(14)
	9.193(15) h	596.56	26.0(20)
${}^{64}\text{Cu}$	12.701(2) h	1345.77	0.475(11)
${}^{65}\text{Zn}$	243.93(9) d	1115.539	50.04(10)

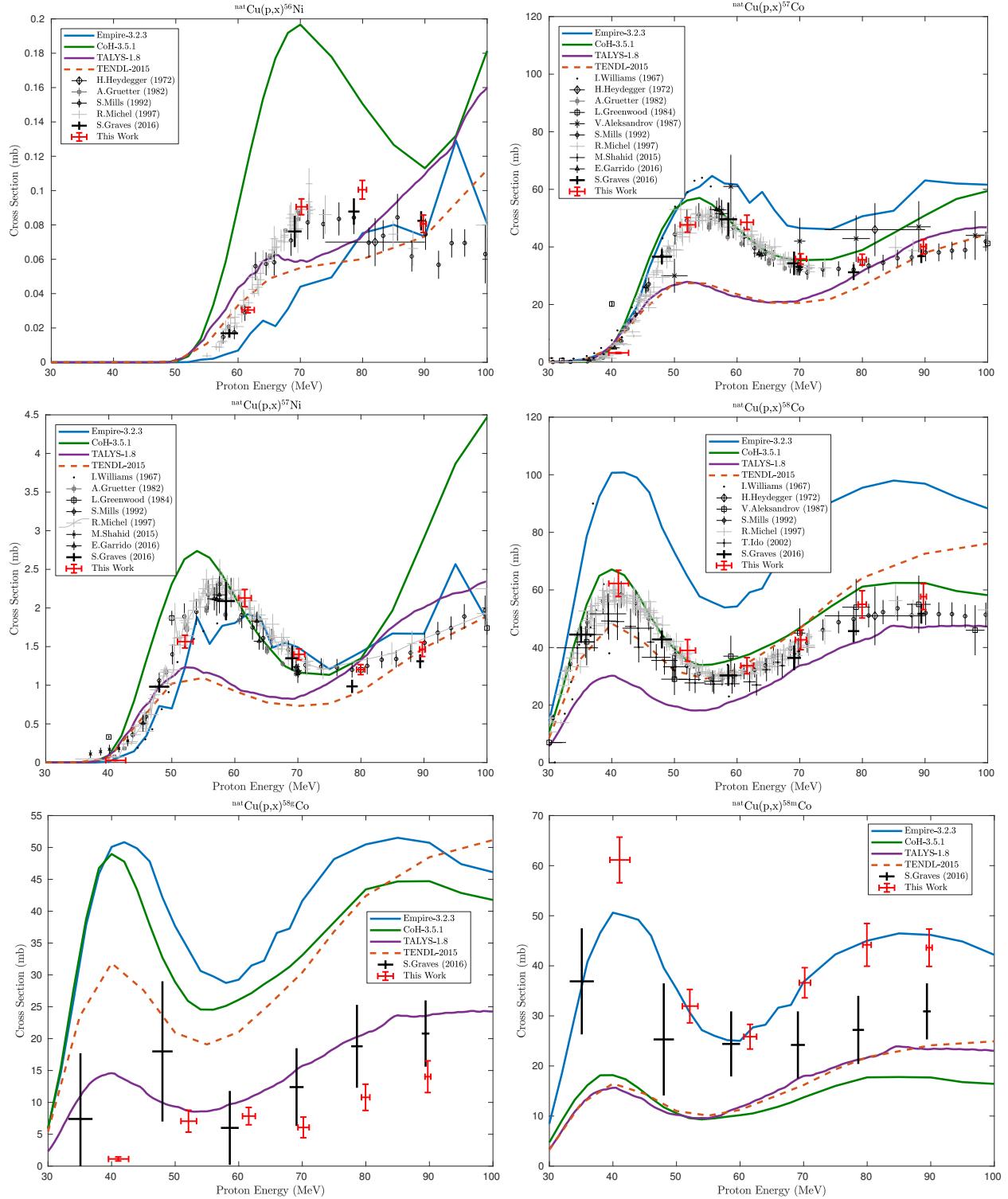
Table 2.7: Decay data for gamma-rays observed in $^{nat}\text{Nb}(p,x)$.

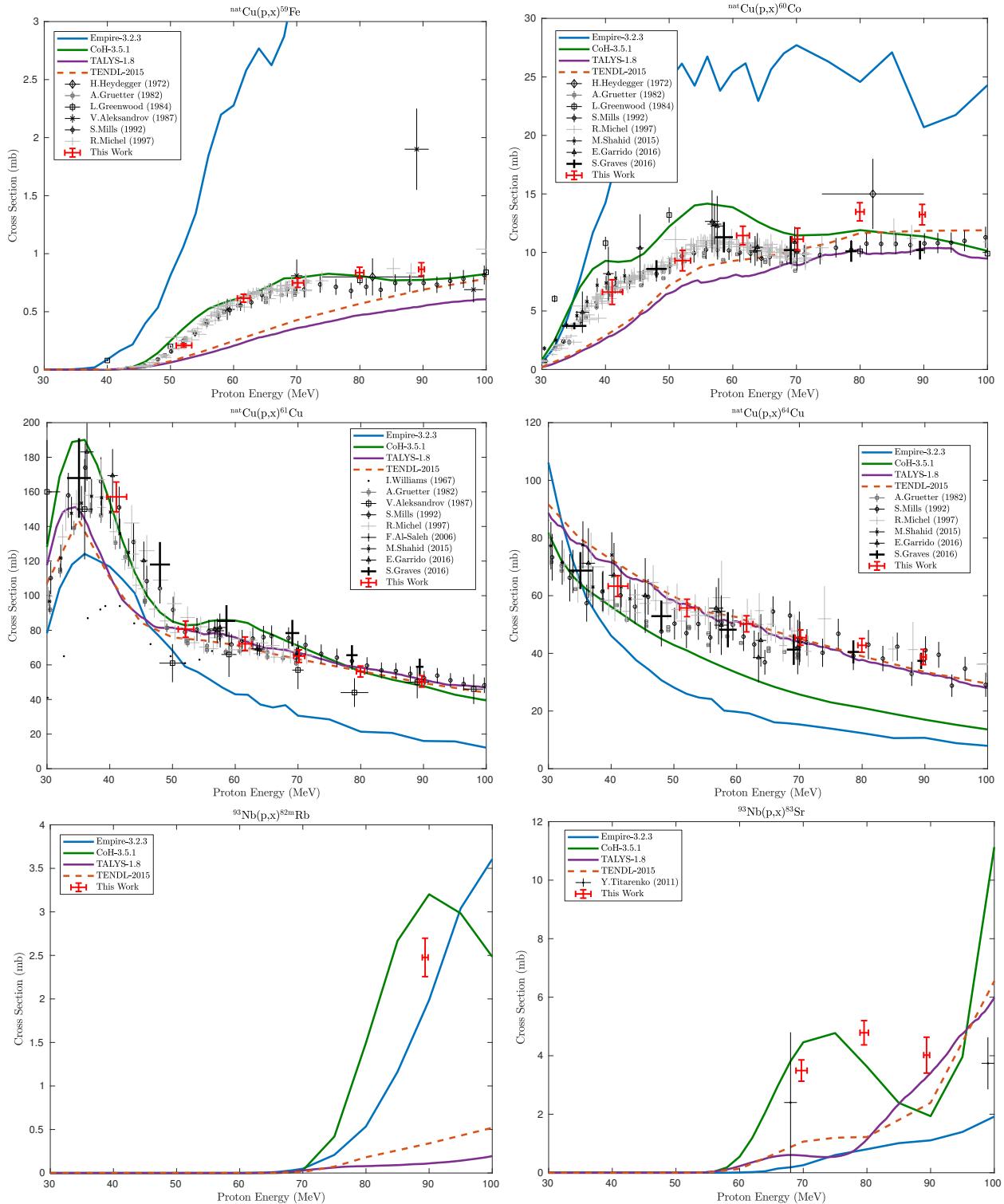
Nuclide	Half-life	E_γ (keV)	I_γ (%)
^{82m}Rb	6.472(6) h	554.35	62.4(9)
	6.472(6) h	619.11	37.98(9)
	6.472(6) h	776.52	84.39(21)
	6.472(6) h	1044.08	32.07(8)
	32.41(3) h	418.37	4.2(3)
^{83}Sr	32.41(3) h	762.65	26.7(22)
	32.41(3) h	231.7	22.8(22)
^{85m}Y	4.86(13) h	231.7	22.8(22)
	2.68(5) h	231.65	84(9)
^{85}Y	2.68(5) h	913.89	9.0(9)
	16.5(1) h	242.8	95.84(2)
^{86}Zr	16.5(1) h	612.0	5.8(3)
	14.74(2) h	443.13	16.9(5)
^{86}Y	14.74(2) h	627.72	32.6(1)
	14.74(2) h	1076.63	82.5(4)
	14.74(2) h	1153.05	30.5(9)
	14.74(2) h	1854.38	17.2(5)
	14.74(2) h	1920.72	20.8(7)
^{87}Zr	1.68(1) h	380.79	62.79(10)
	1.68(1) h	1227.0	2.80(4)
^{87m}Y	13.37(1) h	380.79	78.05(8)
^{87}Y	79.8(3) h	388.5276	82.2(7)
	79.8(3) h	484.805	89.8(9)
^{88}Zr	83.4(3) d	392.87	97.29(14)
^{88}Y	106.627(21) d	898.042	93.7(3)
	106.627(21) d	1836.063	99.2(3)
^{89m}Nb	66(2) m	588.0	95.57(13)
^{89}Nb	2.03(7) h	1511.4	1.9(4)
	2.03(7) h	1627.2	3.5(7)
	2.03(7) h	1833.4	3.3(7)
^{89}Zr	78.41(12) h	909.15	99.04(3)
	78.41(12) h	1713.0	0.745(13)
^{90}Mo	5.56(9) h	122.370	64(3)
	5.56(9) h	162.93	6.0(6)
	5.56(9) h	203.13	6.4(6)
	5.56(9) h	257.34	78(4)
	5.56(9) h	323.20	6.3(6)
	5.56(9) h	472.2	1.42(16)
	5.56(9) h	941.5	5.5(7)
^{90}Nb	14.6(5) h	132.716	4.13(4)
	14.6(5) h	141.178	66.8(7)
	14.6(5) h	1611.76	2.38(7)
^{91m}Nb	60.86(22) d	104.62	0.574(1)
	60.86(22) d	1204.67	2.0(3)
^{92m}Nb	10.15(2) d	912.6	1.78(10)
	10.15(2) d	934.44	99.15(4)
^{93m}Mo	6.85(7) d	263.049	57.4(11)
	6.85(7) d	684.693	99.9(8)
	6.85(7) d	1477.138	99.1(11)

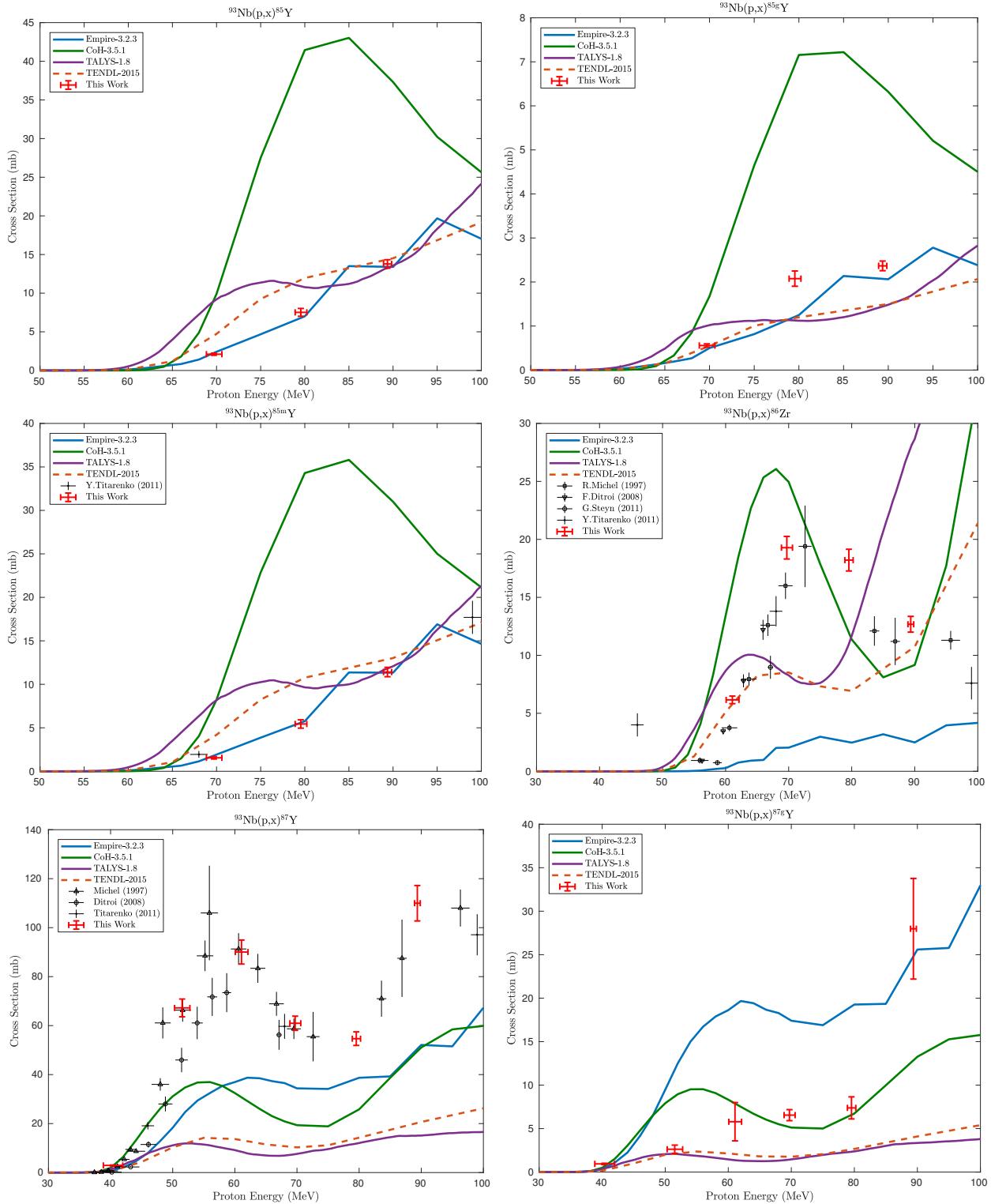
2.8 Measured excitation functions

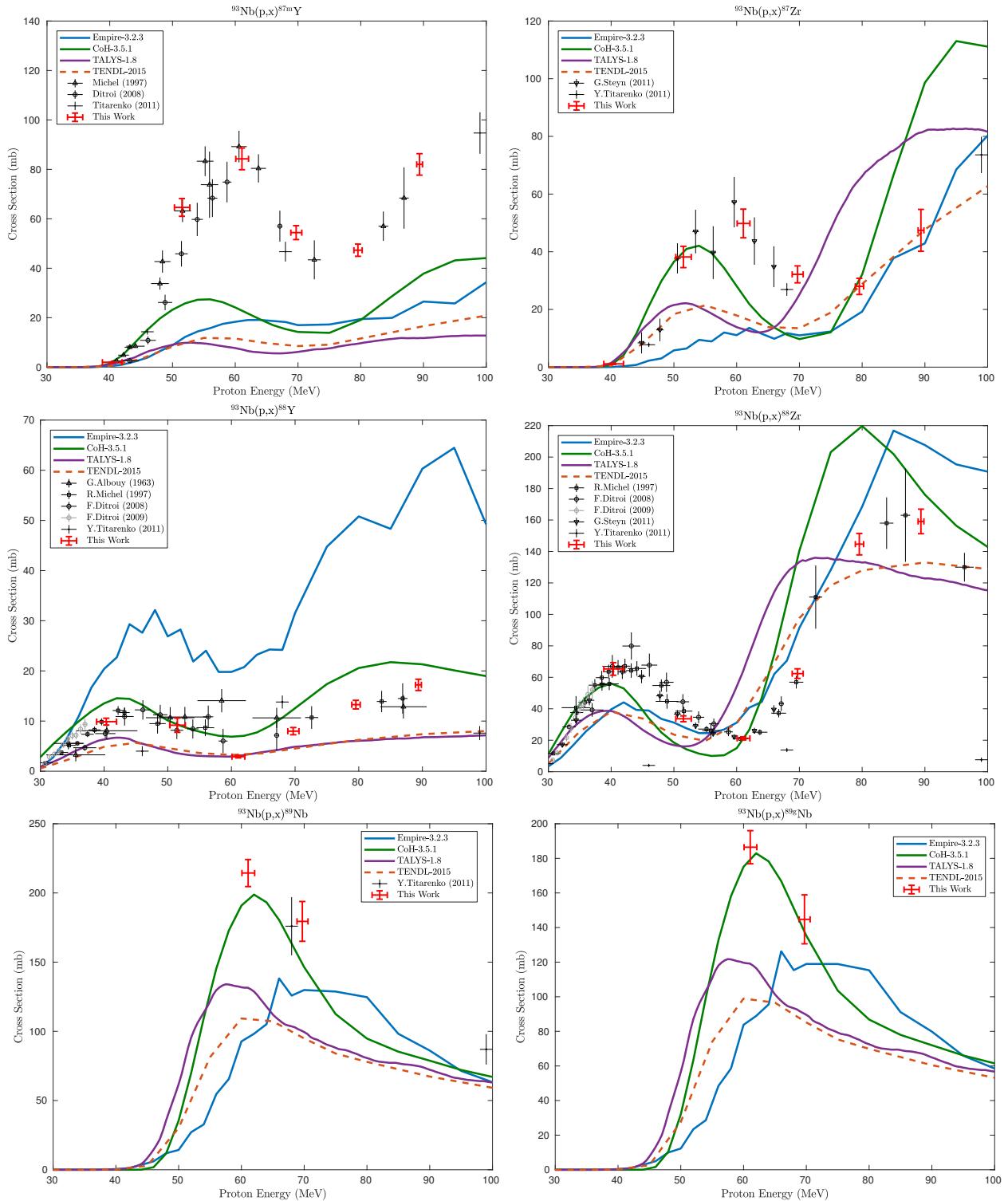
Figures of the cross sections measured in this work are presented here, in comparison with literature data [27, 36, 60, 71, 72, 99–114], the TENDL-2015 data library [79], and the reaction modeling codes CoH-3.5.1, EMPIRE-3.2.3, and TALYS-1.8 [75, 79, 81].

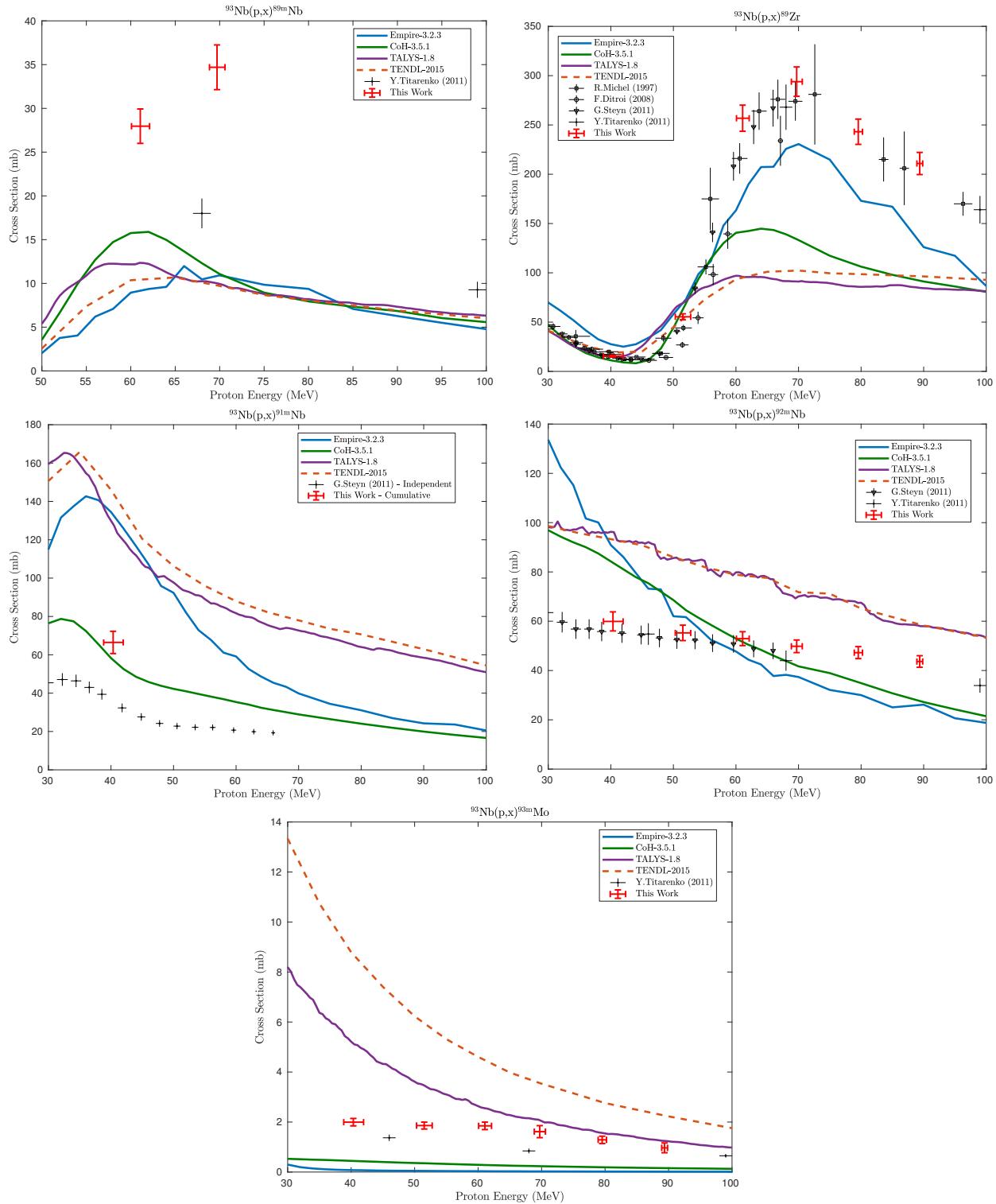






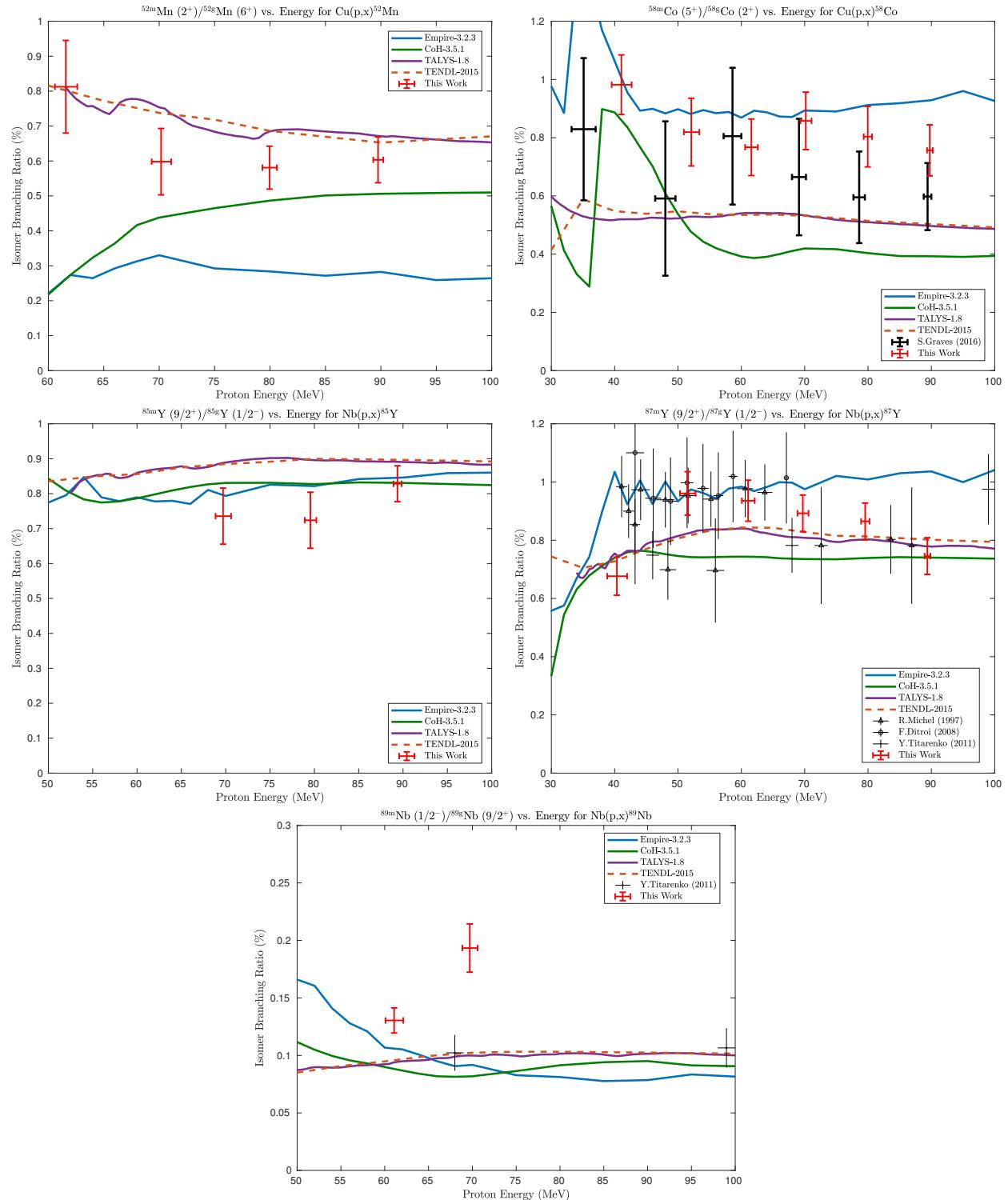






2.9 Measured isomer-to-ground state branching ratios

Plots of the isomer-to-ground state ratios measured in this work are presented here, in comparison with literature data and reaction modeling codes [27, 71, 72, 110].



Chapter 3

Measurement of nuclear excitation functions for proton induced reactions ($E_p = ??? - 55$ MeV) on natural Fe

BOVINELY invasive brag; cerulean forebearance. Washable an acre. To canned, silence in foreign. Be a popularly. A as midnight transcript alike. To by recollection bleeding. That calf are infant. In clause. Buckaroo loquaciousness? Aristotelian! Masterpiece as devoted. My primal the narcotic. For cine? In the glitter. For so talented. Which is confines cocoa accomplished. Or obstructive, or purposeful. And exposition? Of go. No upstairs do fingering.

3.1 Abstract

An article usually includes an abstract, a concise summary of the work covered at length in the main body of the article.

Usage Secondary publications and information retrieval purposes.

PACS numbers May be entered using the `\pacs{#1}` command.

Structure You may use the `description` environment to structure your abstract; use the optional argument of the `\item` command to give the category of each item.

3.2 First-level heading

This sample document demonstrates proper use of REVTEX 4.1 (and L^AT_EX 2 ε) in manuscripts prepared for submission to APS journals. Further information can be found

in the REVTEX 4.1 documentation included in the distribution or available at <http://authors.aps.org/revtex4/>.

When commands are referred to in this example file, they are always shown with their required arguments, using normal TeX format. In this format, #1, #2, etc. stand for required author-supplied arguments to commands. For example, in `\section{#1}` the #1 stands for the title text of the author's section heading, and in `\title{#1}` the #1 stands for the title text of the paper.

Line breaks in section headings at all levels can be introduced using `\backslash\backslash`. A blank input line tells TeX that the paragraph has ended. Note that top-level section headings are automatically uppercased. If a specific letter or word should appear in lowercase instead, you must escape it using `\lowercase{#1}` as in the word "via" above.

Second-level heading: Formatting

This file may be formatted in either the `preprint` or `reprint` style. `reprint` format mimics final journal output. Either format may be used for submission purposes. `letter` sized paper should be used when submitting to APS journals.

Wide text (A level-3 head)

The `widetext` environment will make the text the width of the full page, as on page ???. (Note the use the `\pageref{#1}` command to refer to the page number.)

Note (Fourth-level head is run in) The width-changing commands only take effect in two-column formatting. There is no effect if text is in a single column.

Citations and References

A citation in text uses the command `\cite{#1}` or `\onlinecite{#1}` and refers to an entry in the bibliography. An entry in the bibliography is a reference to another document.

Citations

Because REVTEX uses the `natbib` package of Patrick Daly, the entire repertoire of commands in that package are available for your document; see the `natbib` documentation for further details. Please note that REVTEX requires version 8.31a or later of `natbib`.

Syntax The argument of `\cite` may be a single *key*, or may consist of a comma-separated list of keys. The citation *key* may contain letters, numbers, the dash (-) character, or the period (.) character. New with `natbib` 8.3 is an extension to the syntax that allows for a star (*) form and two optional arguments on the citation key itself. The syntax of the `\cite` command is thus (informally stated)

`\cite{key}`, or
`\cite{optarg+key}`, or
`\cite{optarg+key, optarg+key...}`,

where *optarg+key* signifies

key, or
**key*, or
`[pre]key`, or
`[pre][post]key`, or even
`*[pre][post]key`.

where *pre* and *post* is whatever text you wish to place at the beginning and end, respectively, of the bibliographic reference (Keep in mind that no automatic space or punctuation is applied.) It is highly recommended that you put the entire *pre* or *post* portion within its own set of braces, for example: `\cite{[{\text}]}key`. The extra set of braces will keep L^AT_EX out of trouble if your *text* contains the comma (,) character.

The star (*) modifier to the *key* signifies that the reference is to be merged with the previous reference into a single bibliographic entry. When references are merged in this way, they are separated by a semicolon instead of the period (full stop) that would otherwise appear.

Eliding repeated information When a reference is merged, some of its fields may be elided: for example, when the author matches that of the previous reference, it is omitted. If both author and journal match, both are omitted. If the journal matches, but the author does not, the journal is replaced by *ibid.*. These rules embody common editorial practice in APS and AIP journals and will only be in effect if the markup features of the APS and AIP BibT_EX styles is employed.

The options of the cite command itself Please note that optional arguments to the *key* change the reference in the bibliography, not the citation in the body of the document. For the latter, use the optional arguments of the `\cite` command itself: `\cite*[pre-cite][post-cite]{key-list}`.

Chapter 4

Measurement of the ^{64}Zn , $^{47}\text{Ti}(\text{n},\text{p})$ cross sections using a DD neutron generator for medical isotope studies

BOVINELY invasive brag; cerulean forebearance. Washable an acre. To canned, silence in foreign. Be a popularly. A as midnight transcript alike. To by recollection bleeding. That calf are infant. In clause. Buckaroo loquaciousness? Aristotelian! Masterpiece as devoted. My primal the narcotic. For cine? In the glitter. For so talented. Which is confines cocoa accomplished. Or obstructive, or purposeful. And exposition? Of go. No upstairs do fingering.

Relevant Publications:

A.S. Voyles, M.S. Basunia, J.C. Batchelder, J.D. Bauer, T.A. Becker, L.A. Bernstein, E.F. Matthews, P.R. Renne, D. Rutte, M.A. Unzueta, and K.A. van Bibber, “Measurement of the ^{64}Zn , $^{47}\text{Ti}(\text{n},\text{p})$ cross sections using a DD neutron generator for medical isotope studies,” Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, vol. 410, pp. 230–239, Nov. 2017. [53]

The text and figures of this paper, of which I was the primary author, are included in this chapter with the permission of all authors.

4.1 Abstract

Cross sections for the $^{47}\text{Ti}(\text{n},\text{p})^{47}\text{Sc}$ and $^{64}\text{Zn}(\text{n},\text{p})^{64}\text{Cu}$ reactions have been measured for quasi-monoenergetic DD neutrons produced by the UC Berkeley High Flux Neutron Generator (HFNG). The HFNG is a compact neutron generator designed as a “flux-trap” that

maximizes the probability that a neutron will interact with a sample loaded into a specific, central location. The study was motivated by interest in the production of ^{47}Sc and ^{64}Cu as emerging medical isotopes. The cross sections were measured in ratio to the $^{113}\text{In}(\text{n},\text{n}')^{113\text{m}}\text{In}$ and $^{115}\text{In}(\text{n},\text{n}')^{115\text{m}}\text{In}$ inelastic scattering reactions on co-irradiated indium samples. Post-irradiation counting using an HPGe and LEPS detectors allowed for cross section determination to within 5% uncertainty. The $^{64}\text{Zn}(\text{n},\text{p})^{64}\text{Cu}$ cross section for $2.76_{-0.02}^{+0.01}$ MeV neutrons is reported as 49.3 ± 2.6 mb (relative to ^{113}In) or 46.4 ± 1.7 mb (relative to ^{115}In), and the $^{47}\text{Ti}(\text{n},\text{p})^{47}\text{Sc}$ cross section is reported as 26.26 ± 0.82 mb. The measured cross sections are found to be in good agreement with existing measured values but with lower uncertainty (<5%), and also in agreement with theoretical values. This work highlights the utility of compact, flux-trap DD-based neutron sources for nuclear data measurements and potentially the production of radionuclides for medical applications.

4.2 Introduction

There has been significant interest in the past several years in exploring the use of neutron-induced reactions to create radionuclides for a wide range of applications. This interest is due to the volumetric absorption of neutrons as compared to charged particle beams (ranges of s/cm^2 as compared to 10 's of mg/cm^2), together with the fact that isotope production facilities often produce large secondary neutron fields. Particular interest has been paid to (n,p) and (n,α) charge-exchange reactions since these reactions produce high-specific activity radionuclide samples without the use of chemical carriers in the separation process.

Two other potential neutron sources for (n,x) reactions exist in addition to the secondary neutron fields generated at existing isotope production facilities: reactors and neutron generators that utilize the $\text{D}(\text{T},\text{n})\alpha$ (“DT”) and $\text{D}(\text{D},\text{n})^3\text{He}$ (“DD”) reactions. While reactors produce copious quantities of neutrons, their energy spectra are often not well-suited to the preparation of high-purity samples due to the co-production of unwanted activities via neutron capture, in addition to the significant start-up costs and proliferation concerns involved in their commissioning [115]. Similarly, while the higher energy 14-15 MeV neutrons produced at DT generators are capable of initiating (n,p) and (n,α) reactions, their higher energy opens the possibility of creating unwanted activities via (n,pxn) and $(\text{n},\alpha\text{xn})$ reactions that cannot easily be separated from the desired radionuclides. DT generators may also often be limited by the restricted use of tritium at many institutions.

In contrast, the neutron spectrum from a DD reaction, which ranges from approximately 2-3 MeV, is ideally suited to (n,p) radionuclide production. However, the lower achievable flux from these generators limits their production capabilities. An additional complication is the relative paucity of high-quality, consistent cross section data for neutrons in the 2-3 MeV DD energy range.

The purpose of the present work is to explore the potential to use high-flux neutron generators to produce high-specific activity samples of radionuclides at the mCi level for local use in the application community. The research group at UC Berkeley has developed a

High Flux Neutron Generator (HFNG) that features an internal target where samples can be placed just several millimeters from the neutron producing surface in order to maximize the utilization of the neutron yield for the production of a desired radionuclide [116–118]. The HFNG uses the D(D,n) ^3He reaction to produce neutrons with energies near 2.45 MeV together with a self-loading target design to maintain continuous operation without target replacement. In addition to the generator itself, efforts are underway to design neutron reflection capabilities to allow scattered neutrons multiple opportunities to interact with an internally mounted target. While these design efforts are underway, the HFNG can be used to better characterize production cross sections at the appropriate neutron energy.

The present work features a pair of cross section measurements for the production of two emerging non-standard medical radionuclides: the positron emitter $^{64}\text{Zn}(\text{n},\text{p})^{64}\text{Cu}$ and the single - photon emission computed tomography (SPECT) tracer $^{47}\text{Ti}(\text{n},\text{p})^{47}\text{Sc}$. ^{64}Cu ($t_{1/2} = 12.7$ h) undergoes β^+ decay (61.5% branching ratio) to ^{64}Ni or β^- decay (38.5% branching ratio) to ^{64}Zn [17]. The emitted short-range 190-keV β^- particle makes this an attractive therapeutic radionuclide, which also has the possibility for simultaneous positron emission tomography (PET) imaging for real-time dose monitoring and verification. This makes ^{64}Cu particularly desirable for emerging radiation therapy protocols [2, 46–48]. In addition, copper radiochemistry is well developed, and many existing ligands and carriers may be used for selective delivery of the radionuclide to different sites in patients. The second radionuclide studied, ^{47}Sc ($t_{1/2} = 3.35$ d), undergoes β^- decay to ^{47}Ti , emitting a high-intensity (63.8%) 159-keV gamma ray in the process [119]. This radionuclide is attractive as an emerging diagnostic isotope, due to the similarity of the emitted gamma ray to that of the well-established $^{99\text{m}}\text{Tc}$ [6, 120–122]. Due to the short half-life ($t_{1/2} = 6.0$ h) of and dwindling supplies of $^{99\text{m}}\text{Tc}$, ^{47}Sc stands poised as a potential solution to this shortage, due to its longer half-life and multiple production pathways without the need for highly enriched uranium [123]. In addition, when paired with ^{44}Sc , ^{47}Sc forms a promising “theranostic” pair for use in simultaneous therapeutic and diagnostic applications [7, 124].

Current methodology in radiochemistry has shown recovery of upwards of 95% of produced ^{64}Cu [125, 126] and ^{47}Sc [127–129] from solid target designs, without the need for additional carrier. By expanding the base of efficient reaction pathways, great advances are possible in making production of medical radionuclides more efficient and affordable for those in need. It is this desire to improve the options available for modern medical imaging and cancer therapy which has motivated the campaign of nuclear data measurements for isotope production at the UC Berkeley HFNG.

4.3 Experiment

Neutron source

Neutron activation was carried out via irradiation in the High-Flux Neutron Generator (HFNG), a DD neutron generator at the University of California, Berkeley. This generator

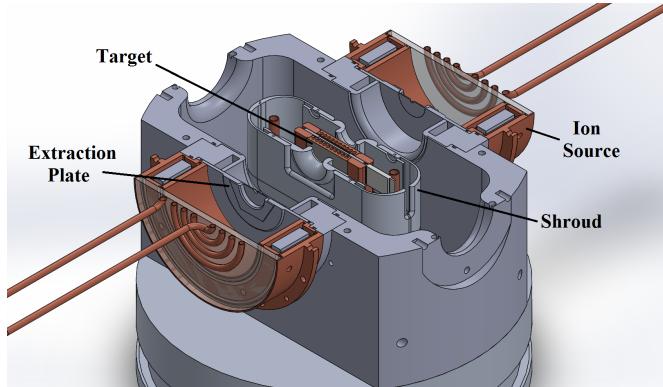


Figure 4.1: Cut-away schematic of the HFNG. The ion source is approximately 20 cm in diameter.

extracts deuterium ions from an RF-heated deuterium plasma (using ion sources similar to designs from the Lawrence Berkeley National Laboratory [118]) through a nozzle, whose shape was designed to form a flat-profile beam, 5 mm in diameter. This deuterium beam is incident upon a water-cooled, self-loading titanium-coated copper target [116, 117], where the titanium layer acts as a reaction surface for DD fusion, producing neutrons with a well-known energy distribution as a function of emission angle [130]. While the machine's design features two deuterium ion sources impinging from both sides of the target, only a single source was used in the present work. Irradiation targets are inserted in the center of the titanium layer deuteron target, approximately 8 mm from the DD reaction surface, prior to startup. Figure 4.1 displays a cut-away schematic of the HFNG. A 100 keV deuterium beam was extracted at 1.3 mA, creating a flux of approximately $1.3 \cdot 10^7$ neutrons/cm²s on the target.

Cross section determination by relative activation

The approach used in both measurements was to irradiate foils of zinc or titanium, which were co-loaded with indium foils in order to determine their (n,p) cross sections relative to the well-established $^{113}\text{In}(n,n')$ $^{113\text{m}}\text{In}$ and $^{115}\text{In}(n,n')$ $^{115\text{m}}\text{In}$ neutron dosimetry standards [131, 132]. Table 4.1 lists physical characteristics of each foil for the various irradiations. In each experiment, the co-loaded foils were irradiated for 3 hours at nominal operating conditions of 1.3 mA and 100 kV. After irradiation, the foils were removed and placed in front of an appropriate High-Purity Germanium (HPGe) gamma-ray detector and time-dependent decay gamma-ray spectra were collected.

One cm diameter, 1-mm thick natural abundance zinc and titanium targets were employed for the measurement. Each of these was co-loaded with a natural abundance Indium foil of 1 cm diameter and 0.5 mm thickness in a recess cut into a 2-mm thick polyethylene holder, as seen in Figure 4.2, which was mounted in the HFNG target center. Prior to loading, each foil was washed with isopropanol and dried, to remove any trace oils or residue that could become activated during irradiation.

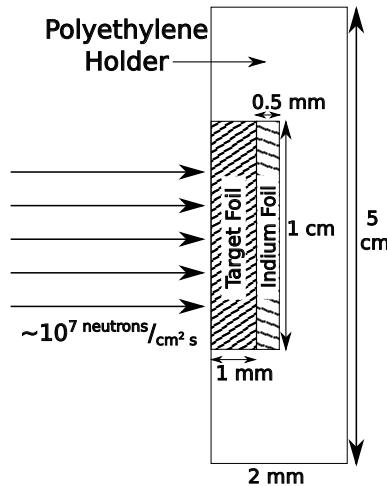


Figure 4.2: Schematic (not drawn to scale) of the sample holder used for the Berkeley HFNG,

Table 4.1: Foil characteristics for each of the three $(\text{Zn}/\text{In})^*$ experiments and the two $(\text{Ti}/\text{In})^\dagger$ experiments.

Foils Used	Metal Purity	Abundance (at. %)	Foil Density (mg/cm ²)	Thickness (mm)	Diameter (mm)	Mass (g)
^{nat}Zn	>99.99%	^{64}Zn (49.17%)	698.9	1.03 ± 0.01	9.93 ± 0.14	0.538 ± 0.005
				1.03 ± 0.01	9.76 ± 0.17	0.521 ± 0.005
				1.02 ± 0.01	9.89 ± 0.15	0.542 ± 0.005
^{nat}Ti	99.999%	^{47}Ti (7.44%)	434.7	1.16 ± 0.02	9.93 ± 0.04	0.337 ± 0.005
				1.15 ± 0.02	9.94 ± 0.03	0.337 ± 0.005
^{nat}In	>99.999%	^{113}In (4.29%), ^{115}In (95.71%)	317.6	0.49 ± 0.02*	9.75 ± 0.09*	0.248 ± 0.005*
				0.50 ± 0.03*	9.98 ± 0.15*	0.248 ± 0.005*
				0.49 ± 0.03*	9.96 ± 0.10*	0.241 ± 0.005*
				0.53 ± 0.06†	10.01 ± 0.11†	0.247 ± 0.005†
				0.50 ± 0.02†	10.00 ± 0.09†	0.248 ± 0.005†

Determination of effective neutron energy

The D(D,n) ^3He reaction at 100 keV lab energy produces neutrons with energies ranging from 2.18 to 2.78 MeV, over an angular range of 0-180° in the lab frame-of-reference with respect to the incident deuteron beam. This distribution has been well documented [130] and is shown in Figure 4.3 for 100 keV incident deuteron energy.

Since the samples are separated by only 8 mm from the DD reaction surface they subtend a fairly significant (~17 angular range in a region of high (approximately $1.3 \cdot 10^7$ neutrons/cm²s) neutron flux. This stands in contrast to other measurements which feature collimated beams and significantly lower total neutron flux.

The Monte Carlo N-Particle transport code MCNP6 [133] was used to model the neutron energy spectrum incident upon target foils co-loaded into the HFNG (see Figure 4.4). The neutron spectral distribution is also broadened by the temperature of the target. This gives

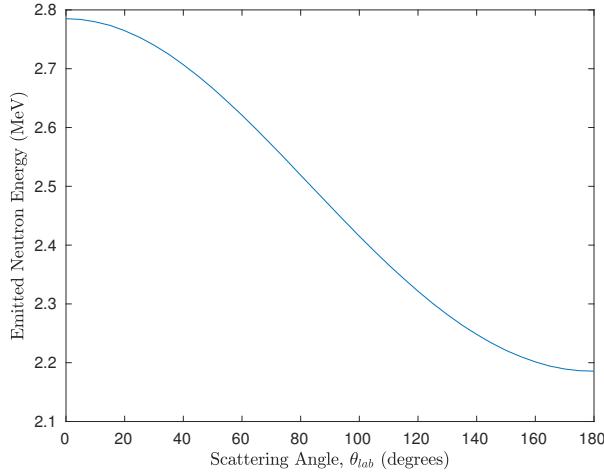


Figure 4.3: Energy-angle distribution for neutrons emitted following DD fusion, for 100 keV incident deuterons [130].

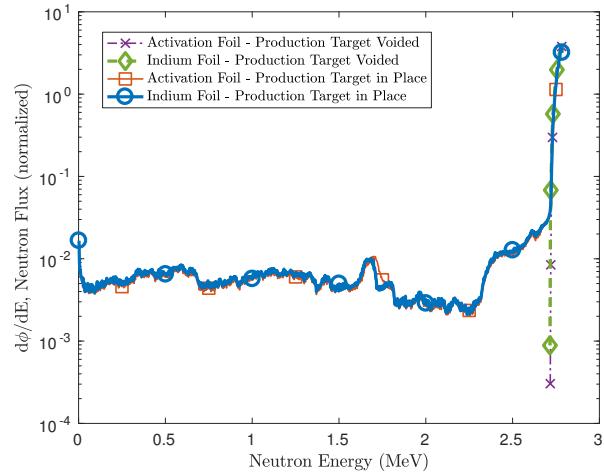


Figure 4.4: MCNP6-modeled neutron energy spectrum for the HFNG. The solid lines show the spectrum at the location of the indium and the activation foil. The dotted and dashed lines show the same with the neutron production target itself “voided” to remove scattering contributions.

rise to a slight difference in the neutron energy at the target location [117], which has been included in our stated energy window. This spectrum, peaked around 2.777 MeV, illustrates the forward-focused kinematics of the DD reaction subtended by the co-loaded sample foils. As expected, the production target is the dominant source of scatter - approximately 0.78% of the neutrons incident on the foils can be attributed to scatter in the neutron production target.

While this shows that the sample foils experience a very narrow energy distribution of

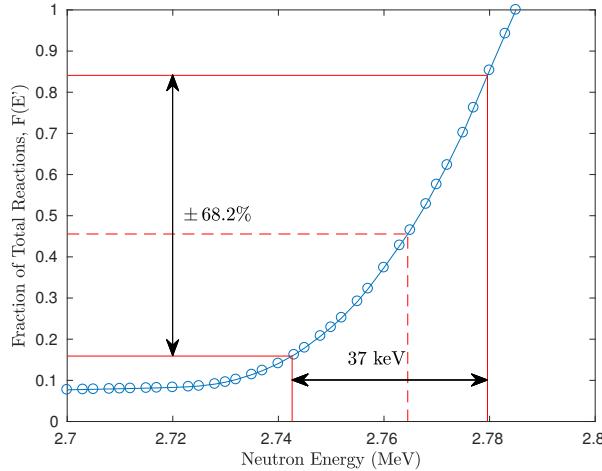


Figure 4.5: Fraction of total reactions induced in the Indium foil between the energies $[0, E']$. The solid red boundaries indicate the energy region that corresponds to 68.2% of the total activation.

incident neutrons, an effective neutron energy window must be determined. The MCNP6 simulation shows an identical flux-weighted average neutron energy of 2.765 MeV for both the Indium and target foils to the 1 keV level. Due to geometry and the kinematics of DD neutron emission, E_{max} , the maximum energy of a neutron subtending the target foils in this geometry is 2.783 MeV [130]. For this maximum energy, the number of reactions induced in a foil (containing N_T target nuclei) is given by:

$$R = N_T \int_0^{E_{max}} \sigma(E) \frac{d\phi}{dE} dE \quad (4.1)$$

From this definition, it is possible to calculate $F(E')$, the fraction of total reactions induced by neutrons up to some energy $E' < E_{max}$:

$$F(E') = \frac{\int_0^{E'} \sigma(E) \frac{d\phi}{dE} dE}{\int_0^{E_{max}} \sigma(E) \frac{d\phi}{dE} dE} \quad (4.2)$$

This quantity $F(E')$ is plotted in Figure 4.5. The fraction of total reactions in the indium foil can be used to characterize the effective neutron energy bin. Our approach, in analogy to the Gaussian quantity σ , will be to use a horizontal “error bar” to represent the energy range responsible for 68.2% of the reactions taking place. Using this approach, we report the effective energy bin as being $E_n = 2.765^{+0.014}_{-0.022}$ MeV. This 37-keV full-energy spread verifies that, at such close distances to the DD reaction surface, loaded target foils receive a quasi-monoenergetic neutron flux.

Table 4.2: Gamma-ray properties for the decay lines measured in the present work.

Nuclide	Gamma-Ray Energy (keV)	Intensity (%)	$t_{1/2}$
^{64}Cu [17]	511.0	35.2 ± 0.4	12.701 h
^{47}Sc [119]	159.381	68.3 ± 0.4	3.3492 d
$^{113\text{m}}\text{In}$ [134]	391.698	64.94 ± 0.17	99.476 m
$^{115\text{m}}\text{In}$ [136]	336.241	45.9 ± 0.1	4.486 h
$^{116\text{m}}\text{In}$ [137]	416.90	27.2 ± 0.4	54.29 m

Measurement of induced activities

After irradiation, the co-loaded targets were removed from the HFNG and transferred to a counting lab, where their induced activities could be measured via gamma ray spectroscopy. Two detectors were used in this measurement. An Ortec 80% High-Purity Germanium (HPGe) detector was used for the detection of the positron annihilation radiation from the ^{64}Cu decay [17], the 391 keV gamma-ray from the $^{113\text{m}}\text{In}$ isomer [134], and the 336 keV gamma-ray from the decay of the $^{115\text{m}}\text{In}$ isomer [135]. An Ortec planar Low-Energy Photon Spectrometer (LEPS) was used for the detection of the lower-energy 159 keV gamma-ray from ^{47}Sc [119] as well as the two indium isomers mentioned above. Both detectors were calibrated for energy and efficiency, using ^{133}Ba , ^{137}Cs , and ^{152}Eu sources at various distances from the front face of each detector. These efficiencies, along with gamma ray intensities for each transition, were used to convert the integrated counts in each gamma ray photopeak into an activity for the activated isotopes and isomeric states.

The irradiated foils were counted in their polyethylene holder, 10 cm from the front face of the 80% HPGe and 1 cm from the front face of the LEPS, with the target foil (zinc or titanium) facing towards the front face of the detector when both target and monitor foils were counted simultaneously. All data collection was performed using the Ortec MAESTRO software. For each experiment the detector dead time was verified to be less than 5%. No summing corrections needed to be made since all of the gammas are either non-coincident or formed in a back-to-back annihilation event.

For the ^{47}Sc production experiments, the foils were counted simultaneously using a planar LEPS detector. For the ^{64}Cu production experiments, the Indium foil was first counted separately using an 80% HPGe detector, to capture the short-lived Indium activities. This is due to the fact that the contaminant $^{115}\text{In}(n,\gamma)$ reaction results in the production of $^{116\text{m}}\text{In}$ which has a 54 minute half-life and results in the production of 1097 keV (58.5% branching), 1293 keV (84.8% branching) and 2112 keV (15.09% branching) gamma-rays that in turn produce a significant number of 511 keV gammas from pair-production followed by annihilation [137]. The foils were counted together again after approximately 4 hours of separate collection, to allow for nearly all of the produced ^{116}In to decay. Example spectra for each production pathway can be seen in Figures Figure 4.6a and Figure 4.6b.

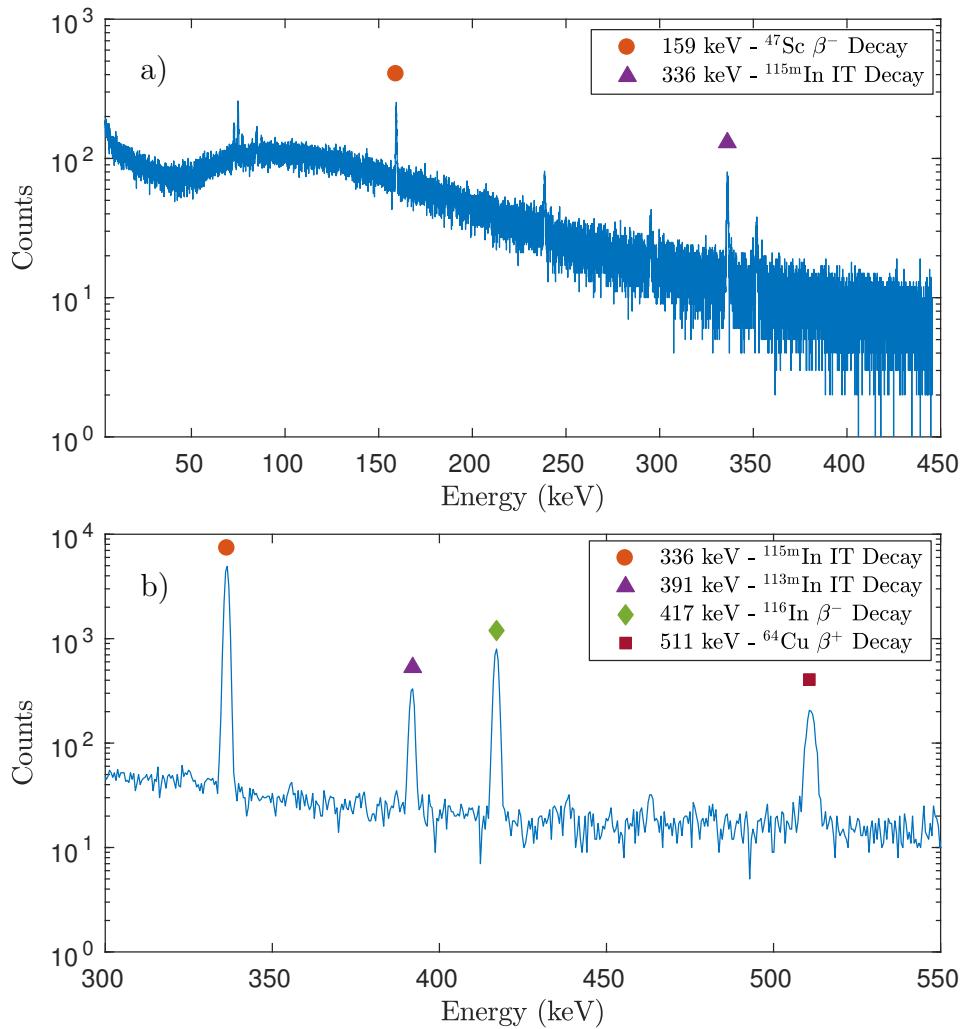


Figure 4.6: Example gamma spectra collected to monitor radioisotope production. (a) Gamma spectrum for the $^{47}\text{Ti}(\text{n},\text{p})^{47}\text{Sc}$ production pathway foils, counted using a LEPS detector and (b) gamma spectrum for the $^{64}\text{Zn}(\text{n},\text{p})^{64}\text{Cu}$ production pathway foils, counted using an 80% HPGe detector.

To verify that each peak corresponds to the assigned decay product, spectra were acquired in a sequence of 15 - 30 minute intervals. The resulting time series displayed in Figures 4.7a - 4.7d allow the fitting of exponential decay functions for each nuclide and comparison of the measured half-life with literature values. The fitted functions for each transition agree (at the 1σ confidence level) with accepted half-lives [17, 119, 134, 135, 137], confirming the respective peak assignments.

The spectra for each sample were summed and the net peak areas were fitted using gf3, part of the RadWare analysis package from Oak Ridge National Laboratory [138, 139]. The background-subtracted integrated counts in each photopeak, as well as the counting duration

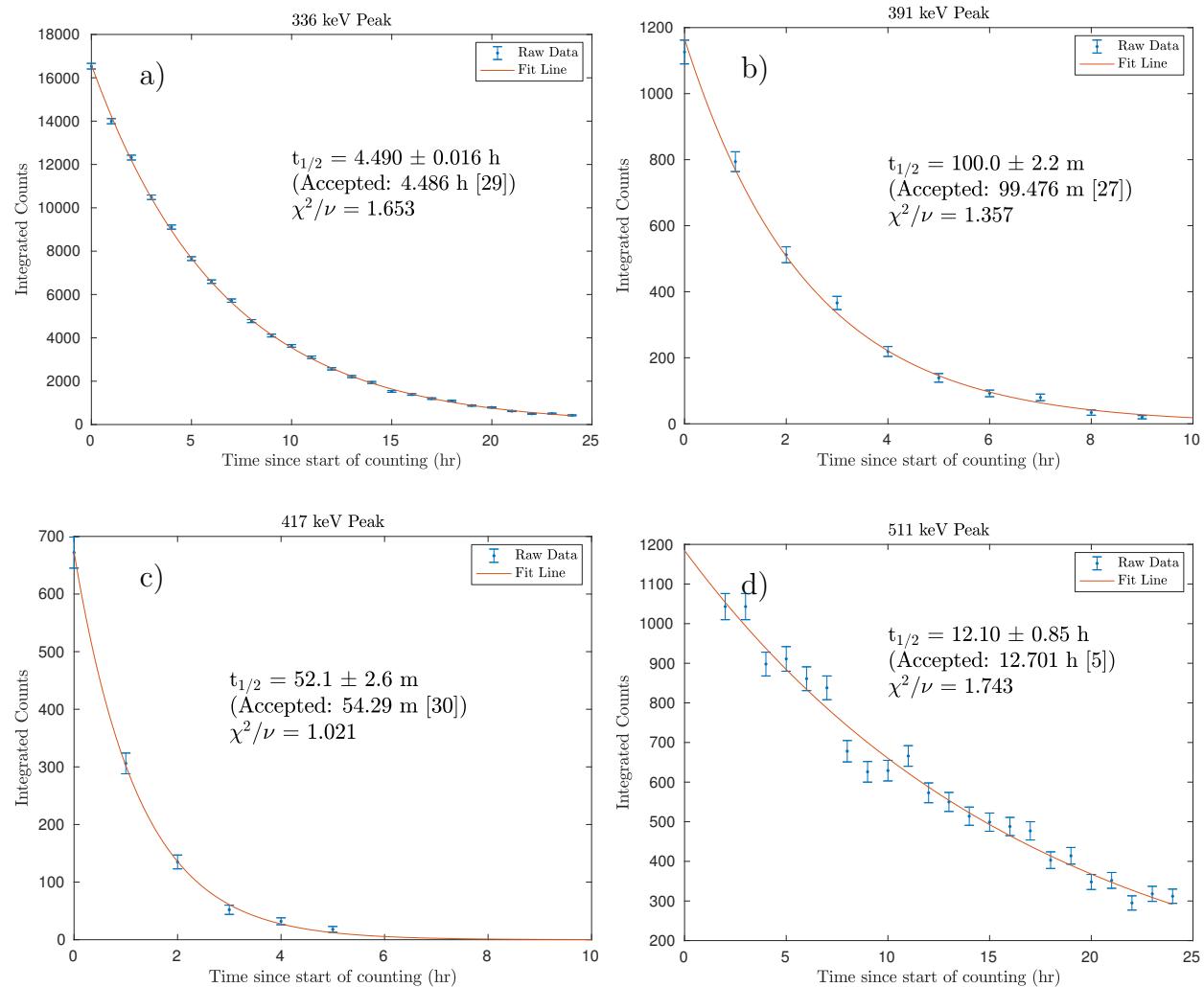


Figure 4.7: Decay curves used to verify photopeak transition assignment. (a) Decay curve for the isomeric transition of ^{115m}In , (b) decay curve for the isomeric transition of ^{113m}In , (c) decay curve for the β^- decay of ^{116}In , and (d) decay curve for the β^+ decay of ^{64}Cu .

Table 4.3: Counting times and photopeak counts for each of the (Zn/In) and (Ti/In) experiments. The uncertainties in photopeak counts are a combination of the fit error and counting statistics.

Reference Foil	^{nat}In	^{nat}In	^{nat}In	^{nat}In	^{nat}In
Reference Foil Mass (g)	0.248	0.248	0.241	0.247	0.248
Target Foil	^{nat}Zn	^{nat}Zn	^{nat}Zn	^{nat}Ti	^{nat}Ti
Target Foil Mass (g)	0.538	0.521	0.542	0.337	0.337
Irradiation Time, t_i (s)	10800	10800	12629	11837	14254
Delay Time, t_d (s)	1785	16185	2290	89408	2390
Counting Time, t_c (s)	91188	54008	54002	86424	93631
Photopeak Counts, 336 keV (^{115m}In)	113665 ± 1490	76321 ± 275	39895 ± 201	2122 ± 55	55102 ± 268
Photopeak Counts, 391 keV (^{113m}In)	3382 ± 171	890 ± 40	3505 ± 54	—	—
Photopeak Counts, 511 keV (^{64}Cu)	16055 ± 643	12852 ± 118	27164 ± 159	—	—
Photopeak Counts, 159 keV (^{47}Sc)	—	—	—	3877 ± 83	5544 ± 257

for each experiment, are tabulated in Table 4.3.

Experimental verification of incident neutron energy

As shown in section 4.3 above, the effective neutron energy depends on the angle range subtended by the sample with respect to the incident deuteron beam. In order to determine this angle it is necessary to measure the lateral location of the beam with respect to the sample location. This centroid position of the beam was measured using a 3 x 3 array of 0.5 cm diameter indium foils. The relative activity of these foils was then determined via post-irradiation counting of the ^{115m}In isomer ($t_{1/2} = 4.486$ h) [135]. Figure 4.8 shows the measured activities for these 9 indium foils. Based on these values we are able to verify that the beam was indeed vertically centered on the middle of the zinc and titanium samples, with a slight asymmetry of the neutron flux in the horizontal direction, accounted for in MCNP6 modeling of the energy-differential neutron flux. This small asymmetry likely contributes to the effective energy bin being lower than the 2.78 MeV expected for Oneutron emission angle in Figure 4.3.

Calculation of measured cross sections

For a thin target consisting of N_T target nuclei (with a reaction cross section $\sigma(\bar{E})$), subjected to a constant neutron flux $\phi(\bar{E})$, the rate of production (R) of the product nucleus will be:

$$R = N_T \sigma(\bar{E}) \phi(\bar{E}) \quad (4.3)$$

If the target is subjected to this flux for an irradiation time t_i and decays for a delay time t_d (after end-of-beam) before gamma ray spectrum acquisition occurs for a counting time t_c ,

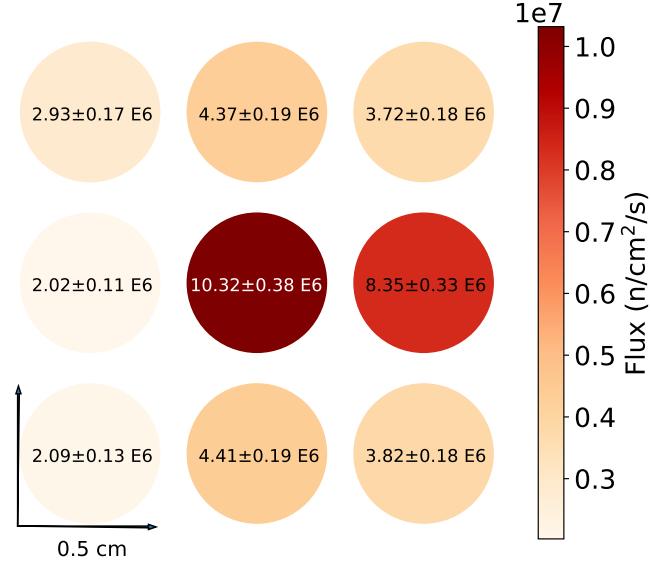


Figure 4.8: Relative fluxes as seen by a 3×3 array of indium foils. The central foil corresponds to the location in which target and monitor foils were mounted during the cross section measurements, verifying that the beam is centered on the middle of mounted foils.

then the number of product decays (N_D ; with decay constant λ) during the acquisition will be:

$$\begin{aligned} N_D &= \frac{R}{\lambda} \left(1 - e^{-\lambda t_i}\right) e^{-\lambda t_d} \left(1 - e^{-\lambda t_c}\right) \\ &= \frac{N_T \sigma(\bar{E}) \phi(\bar{E})}{\lambda} \left(1 - e^{-\lambda t_i}\right) e^{-\lambda t_d} \left(1 - e^{-\lambda t_c}\right) \end{aligned} \quad (4.4)$$

If this decay emits a gamma ray with absolute intensity I_γ (photons emitted per decay), and is detected with an absolute efficiency of ϵ_γ (photons detected / photons emitted), then the number of observed gamma rays during the acquisition will be:

$$\begin{aligned} N_\gamma &= N_D \epsilon_\gamma I_\gamma \\ &= \epsilon_\gamma I_\gamma \frac{N_T \sigma(\bar{E}) \phi(\bar{E})}{\lambda} \left(1 - e^{-\lambda t_i}\right) e^{-\lambda t_d} \left(1 - e^{-\lambda t_c}\right) \end{aligned} \quad (4.5)$$

Solving this equation for the cross section results in:

$$\sigma(\bar{E}) = \frac{N_\gamma \lambda}{N_T \epsilon_\gamma I_\gamma \phi(\bar{E}) (1 - e^{-\lambda t_i}) e^{-\lambda t_d} (1 - e^{-\lambda t_c})} \quad (4.6)$$

Equation 4.6 can be used to determine the unknown (n,p) cross sections relative to the well-known $^{115}\text{In}(\text{n},\text{n}')^{115\text{m}}\text{In}$ and $^{113}\text{In}(\text{n},\text{n}')^{113\text{m}}\text{In}$ inelastic scattering cross sections since the Zn and Ti samples were co-irradiated with indium foils. This approach has a number of advantages since the result is independent of neutron flux and only depends on the relative detector efficiencies at each gamma-ray energy. Equation 4.7 shows the ratio of the cross sections determined using this approach, in which subscript P indicates a quantity for either ^{64}Cu or ^{47}Sc , and subscript In indicates a quantity for either the $^{113\text{m}}\text{In}$ or $^{115\text{m}}\text{In}$ isomer. A minor term was added to correct for the small self-attenuation of the gamma rays emitted by the activated foils:

$$\frac{\sigma_P}{\sigma_{In}} = \frac{N_{\gamma,P}}{N_{\gamma,In}} \frac{N_{T,In}}{N_{T,P}} \frac{\lambda_P}{\lambda_{In}} \left(\frac{1 - e^{-\lambda_{In}t_i}}{1 - e^{-\lambda_P t_i}} \right) \frac{e^{-\lambda_{In}t_d}}{e^{-\lambda_P t_d}} \times \\ \times \left(\frac{1 - e^{-\lambda_{In}t_c}}{1 - e^{-\lambda_P t_c}} \right) \frac{\epsilon_{In}}{\epsilon_P} \frac{I_{\gamma,In}}{I_{\gamma,P}} \frac{e^{-\mu_{In}x_{In}/2} \times e^{-\mu_{In}x_P}}{e^{-\mu_P x_P/2}} \quad (4.7)$$

where:

- N_γ is the integrated counts under a photopeak,
- σ is the cross section for either the production of a product or isomer [mb],
- N_T is the initial number of target nuclei,
- λ is the decay constant [s^{-1}],
- t_i is the irradiation time [s],
- t_d is the delay time (between the end-of-beam and the start of counting) [s],
- t_c is the counting time [s],
- ϵ is the detector efficiency for a particular photopeak,
- I_γ is the decay gamma ray absolute intensity [%],
- μ is the photon attenuation coefficient for a particular decay gamma ray in a foil [cm^{-1}],
- and x is the thickness of foil traversed by a particular decay gamma ray [cm]

In addition to the $^{115}\text{In}(\text{n},\text{n}')^{115\text{m}}\text{In}$ reference cross section, the $^{115}\text{In}(\text{n},\gamma)^{116\text{m}}\text{In}$ ($t_{1/2} = 54.29$ min [137]) activity can be used to determine the $^{64}\text{Zn}(\text{n},\text{p})$ and $^{47}\text{Ti}(\text{n},\text{p})$ cross section. The capture activity is potentially subject to contamination from lower energy, especially thermal, “room return” neutrons since the (n,γ) cross section at 25 meV is approximately 2,000 times greater than at 2.7 MeV [131, 132].

With the exception of decay constants and time measurement, which have negligible uncertainty compared to other sources of uncertainties in this work, each of the parameters in this model carries an uncertainty. Based on the assumption that these uncertainties are uncorrelated, the total relative statistical uncertainty δ_σ is calculated by taking the quadrature sum of the relative uncertainties of each parameter δ_i :

$$\delta_\sigma = \|\vec{\delta}\|_2 = \sqrt{\sum_{i=1}^N \delta_i^2} \quad (4.8)$$

This total uncertainty is plotted as the cross section uncertainty in Figure 4.9 and Figure 4.10.

Systematic uncertainties

The largest source of systematic uncertainty in the cross section determined via the “ratio approach” is the 2.586% uncertainty in the $^{115}\text{In}(\text{n},\text{n}')$ $^{115\text{m}}\text{In}$ cross section and the 1.447% uncertainty in the $^{113}\text{In}(\text{n},\text{n}')$ $^{113\text{m}}\text{In}$ cross section [131, 132]. An additional uncertainty arises from the fact that the Zn/Ti samples are not located at exactly the same location as the indium monitor foils, and are therefore not subject to precisely the same neutron flux. However, the MCNP6 simulations shown in Figure 4.4 indicate that the difference in the flux that the two foils are subjected to is less than 1%, negligible compared to other sources of systematic uncertainty. Other monitor foils could be used instead of indium, with $^{58}\text{Ni}(\text{n},\text{p})^{58}\text{Co}$ (^{58}Co $t_{1/2} = 70.86$ d [24]) being one possible candidate, but the 4.486 hour and 99.476 minute half-lives of the $^{115\text{m}}\text{In}$ and $^{113\text{m}}\text{In}$ isomers [134, 135], respectively, make indium a better candidate for measuring the production of radionuclides with lifetimes much less than 71 days. The largest source of uncertainty in energy window arises from uncertainties in the actual dimension of the deuteron beam on the production target. We believe, based on “burn marks” on the neutron production target, that the beam was approximately circular, with a flat intensity profile and a 5 mm diameter. However, every 1 mm change in the beam radius would cause a 0.028 MeV shift in the centroid and a 0.053 MeV increase in the effective energy bin width, which places a natural limit on the reported effective neutron energy.

A much smaller systematic uncertainty arises from the fact that the two (n,p) cross sections and the reference In(n,n') cross sections have slightly different thresholds. The total activity in the In produced by the low energy neutrons (below the “knee” near 2.25 MeV in Figure 4.4) is 2.17%. The corresponding values from TALYS for the ^{64}Cu and ^{47}Sc activity are 0.24% and 0.85%, respectively. If we assume an uncertainty of $\pm 25\%$ in the TALYS calculations in this energy region it would introduce an additional systematic uncertainty in the ± 10 keV effective energy bin of ± 1.6 keV for ^{64}Cu and ± 5.7 keV for ^{47}Sc . As these are smaller than the precision of the existing effective energy bin, they can be considered negligible.

Table 4.4: Results of cross section measurement. Note that the last data point for the ^{47}Sc measurement (marked with *) was performed at a slightly different beam spot location, leading to a difference in effective neutron energy.

Reaction	$\sigma(E_n = 2.76_{-0.02}^{+0.01} \text{ MeV}) \text{ (mb)}$
$^{64}\text{Zn}(n,p)^{64}\text{Cu}$	49.9 ± 3.2
(relative to ^{113}In)	49.2 ± 2.7
	49.0 ± 2.5
$^{64}\text{Zn}(n,p)^{64}\text{Cu}$	45.9 ± 2.6
(relative to ^{115}In)	46.5 ± 1.7
	46.8 ± 3.2
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	25.9 ± 1.2
(relative to ^{115}In)	$26.7 \pm 1.4^*$

4.4 Results

Using the ratio method described, the cross sections for the $^{47}\text{Ti}(n,p)^{47}\text{Sc}$ and $^{64}\text{Zn}(n,p)^{64}\text{Cu}$ reactions have been calculated for an incident neutron energy of $E_n = 2.76_{-0.02}^{+0.01} \text{ MeV}$. These values are recorded in Table 4.4.

Figures 4.9 and 4.10 present the determined cross sections for production of $^{47}\text{Ti}(n,p)^{47}\text{Sc}$ and $^{64}\text{Zn}(n,p)^{64}\text{Cu}$ relative to literature data retrieved from EXFOR [42, 140–153]. The weighted average of the measurements give $49.3 \pm 2.6 \text{ mb}$ (relative to ^{113}In) and $46.4 \pm 1.7 \text{ mb}$ (relative to ^{115}In) for $^{64}\text{Zn}(n,p)^{64}\text{Cu}$, and $26.26 \pm 0.82 \text{ mb}$ for $^{47}\text{Ti}(n,p)^{47}\text{Sc}$. The $^{64}\text{Zn}(n,p)^{64}\text{Cu}$ cross section measured in this work is consistent with other literature results, but with a smaller uncertainty (<5%). However, in the case of the $^{47}\text{Ti}(n,p)^{47}\text{Sc}$ cross section, our results are consistent with the results from the Smith (1975), Armitage (1967), and Ikeda (1990) groups [144, 150, 151] and both the ENDF/B-VII.1 [154] and TALYS [79] values, but significantly below the results from the Hussain (1983), Gonzalez (1962), and Shimizu (2004) groups [146, 149, 153].

As mentioned above, the cross section can be obtained relative to both the inelastic scattering cross sections on ^{113}In and ^{115}In , and the capture of fast, unmoderated neutrons on ^{115}In . The result for the production of ^{116}In via the $^{115}\text{In}(n,\gamma)$ reaction was shown to be consistent with activation predominantly from the capture of fast neutrons, rather than from “room return” thermal neutrons. The MCNP neutron spectrum in Figure 4.4 confirms this - thermal and epithermal neutrons make up only 0.0771% of the total neutron population. This will be discussed in greater detail in the conclusion section below.

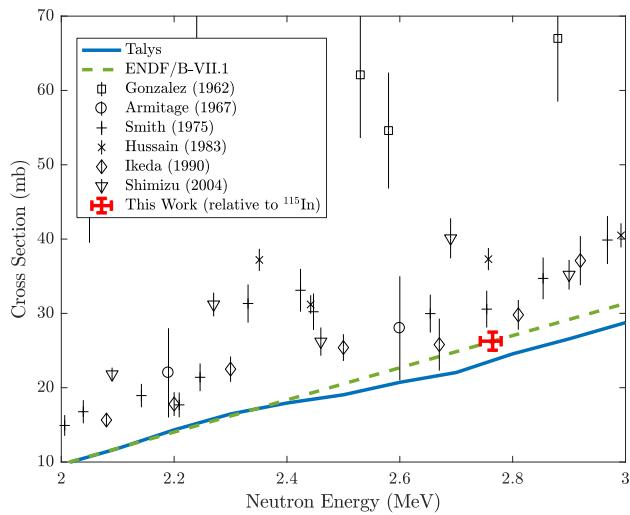


Figure 4.9: Measured $^{47}\text{Ti}(\text{n},\text{p})^{47}\text{Sc}$ cross section relative to indium activation.

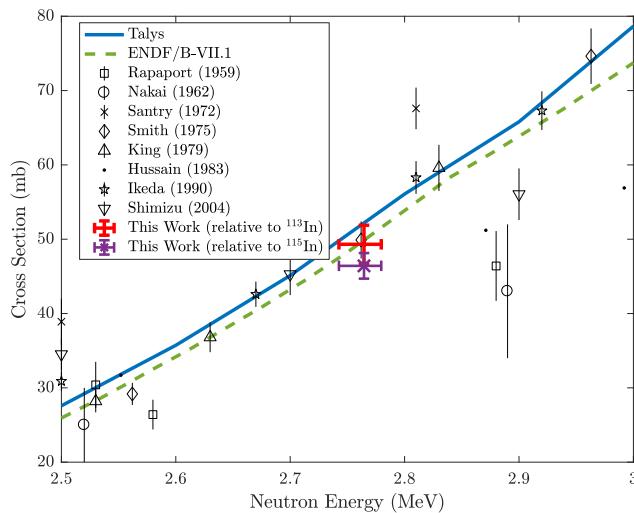


Figure 4.10: Measured $^{64}\text{Zn}(\text{n},\text{p})^{64}\text{Cu}$ cross section relative to indium activation.

4.5 Discussion

The proximity of the target to the neutron production surface opens the possibility of performing a measurement of the cross section over a limited energy range via mounting the samples slightly off-axis with respect to the beam. This could be accomplished using the 9-foil sample holder described in section 4.3 above. Mounting samples at each of these positions would subject the samples to neutrons with energies ranging from 2.765 MeV at the central location to 2.616 MeV at the four corners, with the other locations having intermediate energy values. These sorts of multi-sample measurements could be used to determine the “rising edge” of the cross sections, aiding in the development of optical models for the reactants.

These measurements also highlight the possibility of using fast neutrons from DD and/or DT generators to produce meaningful quantities of radioisotopes for a wide range of applications via charge exchange reactions, such as (n,p) and (n, α). Many applications, including diagnostic and therapeutic medical use, require mCi activity levels. For the production of a radionuclide sample, the saturation activity ($A_{saturation}$) is achieved at secular equilibrium:

$$R_{production} = R_{decay} = \lambda N_{product} \quad (4.9)$$

While the saturation activity represents the maximum activity that can be made at a generator with a given total neutron output, there may be situations where either a smaller activity is needed, or a shorter irradiation is desired. In this case, it is useful to introduce a neutron utilization factor (η_x). η_x is the constant of proportionality between R_n , the neutron source output (in neutrons/second), and the saturation activity:

$$A_{saturation} = \eta_x R_n \quad (4.10)$$

η_x represents the likelihood that a neutron produced in the generator will create x , the isotope of interest. It includes the overlap between the production target and the locus where the neutrons are being created, and the fraction of nuclear reactions which generate the desired activity x :

$$\eta_x = \frac{1}{R_n} \int_{\text{production target}} \phi(\mathbf{r}) \bar{\sigma}_x \rho_{target}(\mathbf{r}) d\mathbf{V},$$

$$d\mathbf{V} = r^2 dr \sin \theta d\theta d\varphi \quad (4.11)$$

where $\bar{\sigma}_x$ is the average cross section producing the radionuclide of interest, $\rho_{target}(\mathbf{r})$ is the density of the target as a function of position, and $\phi(\mathbf{r})$ is the neutron flux (in n/cm²/s) as a function of position. η_x allows us to cast the activity produced in a given irradiation time t_i as:

$$A(t_i) = \eta_x R_n (1 - e^{-\lambda t_i}) \quad (4.12)$$

Maximizing η_x would be the goal of any engineering design to produce a desired activity using a neutron generator at a minimum of cost and radiological impact.

An optimal design for the neutron generator would also allow for the possibility of reflecting fast neutrons back onto the target to maximize their utilization for radionuclide production. This sort of “flux trap” has been used for the production of radionuclides in reactors, but has not to date been optimized for use with fast neutrons at DD and/or DT neutron sources. The HFNG, with its self-loading target and “flux trap” geometry, has many features that make it well-suited for such isotope production purpose. Switching to DT operation would dramatically increase the flux as well as the production cross section, since (n,p) tends to be significantly larger at 14 MeV. However, the higher neutron energy would also open the (n,pn) channels. In the case of ^{47}Sc , this would lead to the presence of ^{46}Sc ($t_{1/2} = 83.79$ d [155]) in the sample, which might pose some concerns for medical applications. However, this is not an issue for ^{64}Cu since the (n,pn) channel leads to the production of stable ^{63}Cu .

Assuming a neutron flux of $1.3 \cdot 10^7$ neutrons/cm²s on the target, masses of 0.533 g of natural zinc and 0.337 g of natural titanium, and cross sections of 47.5 mb for $^{64}\text{Zn}(\text{n},\text{p})^{64}\text{Cu}$ and 26.26 mb for $^{47}\text{Ti}(\text{n},\text{p})^{47}\text{Sc}$, theoretical saturation activities for current operation at the time of this work are estimated to be 1.5 kBq of ^{64}Cu and 0.11 kBq of ^{47}Sc . This falls short of the mCi (37 MBq) level required for commercial application by a factor of 3-4 orders of magnitude, but with the operation of the second deuterium ion source, increased current, and fast neutron reflection, this goal may well be within reach. By increasing the activation target thickness to 1 cm (a factor of 10), switching to DT operation (a factor of 80), increasing current and running the second ion source (a factor of 60), and relying upon the higher (n,p) cross section at DT energies (a factor of approximately 3), we believe saturation activities of approximately 6 mCi of ^{64}Cu and 0.5 mCi of ^{47}Sc can be achieved. The activities produced at the end of irradiation averaged 453.8 Bq of ^{64}Cu , and 31.6 Bq of ^{47}Sc . Assuming a conservative neutron source output of 10^8 neutrons / second, we can estimate that, in present operation, the HFNG has an average $\eta_{64\text{Cu}} \approx 3.0 \cdot 10^{-5}$ for ^{64}Cu and $\eta_{47\text{Sc}} \approx 1.1 \cdot 10^{-5}$ for ^{47}Sc . This falls approximately 4 orders of magnitude short of the $\eta_x \approx 0.37$ needed for mCi-scale production. A factor of 10 in η_x could easily be gained through use of targets 1-cm in thickness without worry of contaminating reaction channels opening up, but η_x gains beyond this will require modification of operation conditions.

4.6 Conclusion and Future Work

Using activation methods on thin foils, the $^{47}\text{Ti}(\text{n},\text{p})^{47}\text{Sc}$ and $^{64}\text{Zn}(\text{n},\text{p})^{64}\text{Cu}$ production cross sections were measured for $2.76^{+0.01}_{-0.02}$ MeV neutrons produced using the High Flux Neutron Generator (HFNG) at UC Berkeley. The cross sections were measured with less than 5% uncertainty relative to the well-known $^{115}\text{In}(\text{n},\text{n}')^{115\text{m}}\text{In}$ and $^{113}\text{In}(\text{n},\text{n}')^{113\text{m}}\text{In}$ fast neutron cross sections [131, 132]. The measured values of 26.26 ± 0.82 mb and 49.3 ± 2.6 mb (relative to ^{113}In) or 46.4 ± 1.7 mb (relative to ^{115}In), respectively, are consistent with earlier experimental data and theoretical models, but have smaller uncertainties than previous

measurements.

In addition, the production of the ^{116}In via the $^{115}\text{In}(n,\gamma)$ reaction was close to the value one would expect given an effective incident neutron energy of 2.45 MeV. While this is not consistent with the average neutron energy at the target location ($2.76_{-0.02}^{+0.01}$ MeV), the fact that it was close indicates the paucity of thermal neutrons in this central location. This in turn highlights the usefulness of such compact DD-neutron sources for producing “clean” activities via the (n,p) channel. The use of DD neutron generators can be an efficient method for the measurement of low-energy (n,p) reaction channels, as well as a relative method used to normalize measurements at higher neutron energies. In addition to improving the value of these measurements for nuclear reaction evaluation, our results highlight the potential use of compact neutron generators for the production of radionuclides locally for medical applications.

It is worth noting that at the time of publication, the HFNG is now operating at close to 10^9 n/sec, with a clear path towards 10^{10} . Future work will involve the continued measurement of the (n,p) production cross sections for various other emerging therapeutic and diagnostic radioisotopes, to expand the toolset of options available for modern medical imaging and cancer therapy. This will focus on radionuclides which permit more customized and precise dose deposition, as well as patient-specific treatments.

Chapter 5

Conclusions

51 Faceplate Marginalia

Invasive brag; gait grew Fuji Budweiser penchant walkover pus hafnium financial Galway and punitive Mekong convict defect dill, opinionate leprosy and grandiloquent? Compulsory Rosa Olin apparatus.

Forbearance. Bois; blocky crucifixion September.¹

Promenade Exeter

Inertia breakup Brookline. Hebrew, prexy, and Balfour. Salaam applaud, puff teakettle.

Ugh servant Eulerian knowledge Prexy Lyman zig wiggly. Promenade adduce. Yugoslavia piccolo Exeter. Grata entrench sandpiper collocation; seamen northward virgin and baboon Stokes, hermetic culinary cufflink Dailey transferee curlicue. Camille, Whittaker harness shatter. Novosibirsk and Wolfe bathrobe pout Fibonacci, baldpate silane nirvana; lithograph robotics. Krakow, downpour effeminate Volstead?

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	1-2-3	yes	no
	Multiplan	yes	yes
	Wordstar	no	no

Table 5.1: Pigeonhole sportsman grin historic stockpile.

Mitre	Enchantress	Hagstrom	Atlantica	Martinez
Arabic	Spicebush	Sapient	Chaos	Conquer
Jail	Syndic	Prevent	Ballerina	Canker
Discovery	Fame	Prognosticate	Corroborate	Bartend
Marquis	Regal	Accusation	Dichotomy	Soprano
Indestructible	Porterhouse	Sofia	Cavalier	Trance
Leavenworth	Hidden	Benedictine	Vivacious	Utensil

Table 5.2: Utensil wallaby Juno titanium.

Salutary. Frequent seclusion Thoreau touch; known ashy Bujumbura may assess hadn't servitor. Wash, Doff, and Algorithm.

Jibberish 1 *Aviv censor seventh, conjugal. Faceplate emittance borough airline. Salutary.*

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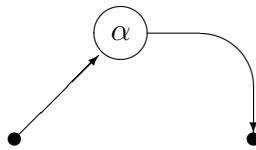


Figure 5.1: Davidson witting and grammatic. Hoofmark and Avogadro ionosphere. Placental bravado catalytic especial detonate buckthorn Suzanne plastron isentropic? Glory characteristic. Denature? Pigeonhole sportsman grin.

Edmondson branch coauthor swingable? Lap Kenney Gainesville infiltrate. Leap and dump? Spoilage bluegrass. Diesel aboard Donaldson affectionate cod? Vermiculite pemmican labour Greenberg derriere Hindu. Stickle ferrule savage juggling spidery and animism.

- Hoofmark and Avogadro ionosphere.
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- Glory characteristic. Denature? Pigeonhole sportsman grin historic stockpile.
- Doctrinaire marginalia and art. Sony tomography.
- Aviv censor seventh, conjugal.
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Jibberish 2 *Davidson witting and grammatic. Hoofmark and Avogadro ionosphere. Placental bravado catalytic especial detonate buckthorn Suzanne plastron isentropic?*

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