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Excitation functions for (p,x) reactions of niobium in the energy range of E_p = 40–90 MeV

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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}Nb(p,4n)^{90}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 ^{nat}Nb(p,x) and ^{nat}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 ^{nat}Cu(p,x)^{56}Co, ^{nat}Cu(p,x)^{62}Zn, and ^{nat}Cu(p,x)^{65}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 ^{nat}Nb(p,x)^{82m}Rb reaction, and of the independent cross sections for ^{nat}Cu(p,x)^{52g}Mn and ^{nat}Nb(p,x)^{85g}Y in the 40–90 MeV region. The effects of ^{nat}Si(p,x)^{22,24}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}Nb(p,x) and ^{nat}Cu(p,x) reactions.

1. 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 ^{22}Na, ^{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 [2,5,6], 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 [6]. 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

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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 for unique identification 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, ε = 100% to ^{48}Ti) and ^{48}Sc ($t_{1/2}$ = 43.67 h, β^- = 100% to ^{48}Ti) can both be formed via ^{nat}Ti(p,x) reactions, yielding the same 983.52 keV transition in ^{48}Ti [7]. 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, and 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}Nb(p,4n)^{90}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 (ε = 100%, $t_{1/2}$ = 5.56 ± 0.09 h [8]) and seven strong, distinct gamma lines (notably its 122.370 keV [I_γ = 64 ± 3%] and 257.34 keV [I_γ = 78 ± 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 ^{nat}Nb(p,x) reaction. In addition to the ^{nat}Nb(p,x)^{90}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 ^{82m}Rb.

In addition to providing a potentially highly-valuable beam monitor, the 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≈190 region [9,10]. These include the ^{52m}Mn ($t_{1/2}$ = 21.1 ± 0.2 m; $J^\pi = 2^+$) to ^{52g}Mn ($t_{1/2}$ = 5.591 ± 0.003 d; $J^\pi = 6^+$), ^{58m}Co ($t_{1/2}$ = 9.10 ± 0.09 h; $J^\pi = 5^+$) to ^{58g}Co ($t_{1/2}$ = 70.86 ± 0.06 d; $J^\pi = 2^+$), ^{85m}Y ($t_{1/2}$ = 4.86 ± 0.13 h; $J^\pi = 9/2^+$) to ^{85g}Y ($t_{1/2}$ = 2.68 ± 0.05 h; $J^\pi = 1/2^-$), ^{87m}Y ($t_{1/2}$ = 13.37 ± 0.03 h; $J^\pi = 9/2^+$) to ^{87g}Y ($t_{1/2}$ = 79.8 ± 0.3 h; $J^\pi = 1/2^-$), and ^{89m}Nb ($t_{1/2}$ = 66 ± 2 m; $J^\pi = 1/2^-$) to ^{89g}Nb ($t_{1/2}$ = 2.03 ± 0.07 h; $J^\pi = 9/2^+$) ratios [11–15].

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. Experimental methods and materials

The work described herein follows the methods established by Graves et al. for monitor reaction characterization of beam energy and fluence in stacked target irradiations [16].

2.1. 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 [17]. A series of nominal 25 μm ^{nat}Nb foils (99.8%, lot #T23A035), 25 μm ^{nat}Al foils (99.999%, lot #M06C032), and 50 μm ^{nat}Cu foils (99.9999%, lot #N26B062) were used as targets (all from Alfa Aesar, Ward Hill, MA, 01835, USA). Six foils of each metal were cut down to 2.5 × 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 3 M 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 Fig. 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

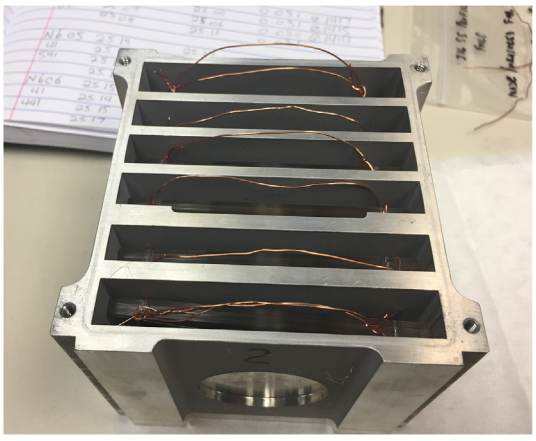

Fig. 1. Photograph of the assembled IPF target stack, before the stack's o-ring lid was sealed in place. The baling wire handles affixed to each bunch of Al + Cu + Nb foils are visible in each energy position, to facilitate removal of activated foils via manipulators in the IPF hot cell. The circular Inconel beam entrance aperture is visible in the bottom center of the photograph.