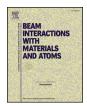
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Excitation functions for (p,x) reactions of niobium in the energy range of $E_p = 40-90 \text{ MeV}$



Andrew S. Voyles^{a,*}, Lee A. Bernstein^{a,b}, Eva R. Birnbaum^c, Jonathan W. Engle^d, Stephen A. Graves^e, Toshihiko Kawano^f, Amanda M. Lewis^a, François M. Nortier^c

- ^a Department of Nuclear Engineering, University of California, Berkeley, Berkeley, CA 94720, USA
- ^b Nuclear Science Division. Lawrence Berkelev National Laboratory. Berkeley. CA 94720. USA
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- ^d Department of Medical Physics, University of Wisconsin Madison, Madison, WI 53705, USA
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ARTICLE INFO

Keywords:

Nb + p

Cu + p Niobium

⁹⁰Mo

Nuclear cross sections Stacked target activation Monitor reactions

Medical isotope production

Isomer branching ratios

MCNP

LANL

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 ⁹³Nb(p,4n) ⁹⁰Mo 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 natNb(p,x) and natCu(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 $^{\text{nat}}\text{Nb}(p,x)^{82m}\text{Rb}$ reaction, and of the independent cross sections for $^{\text{nat}}\text{Cu}(p,x)^{52g}\text{Mn}$ 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 natNb(p,x) and natCu(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 $\,<\,30\,\text{MeV}/\,$ A) and intermediate-energy (30 $\,<\,$ E $\,<\,200\,\text{MeV}/\,$ A) accelerators, e.g., $^{11}\text{C},$ $^{18}\text{F},$ $^{68}\text{Ga},$ $^{82}\text{Rb},$ and $^{123}\text{I}.$ These accelerators also produce non-medical radionuclides with commercial value, such as $^{22}\text{Na},$ $^{73}\text{As},$ $^{95\text{m}}\text{Tc},$ 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 thintarget 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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^{*} Corresponding author.

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Measurement of the ⁶⁴Zn, ⁴⁷Ti(n,p) cross sections using a DD neutron generator for medical isotope studies



A.S. Voyles ^{a,*}, M.S. Basunia ^b, J.C. Batchelder ^a, J.D. Bauer ^c, T.A. Becker ^d, L.A. Bernstein ^{a,b}, E.F. Matthews ^a, P.R. Renne ^{d,e}, D. Rutte ^{d,e}, M.A. Unzueta ^a, K.A. van Bibber ^a

- ^a Department of Nuclear Engineering, University of California, Berkeley, Berkeley, CA 94720, USA
- ^b Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA
- ^c Lawrence Livermore National Laboratory, Livermore, CA 94551, USA
- d Berkeley Geochronology Center, Berkeley, CA 94709, USA
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ARTICLE INFO

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DD neutron generator
Medical isotope production
Scandium (Sc) and copper (Cu)
radioisotopes
Indium
Ratio activation
Theranostics

ABSTRACT

Cross sections for the $^{47}\text{Ti}(n,p)^{47}\text{Sc}$ and $^{64}\text{Zn}(n,p)^{64}\text{Cu}$ reactions have been measured for quasimonoenergetic 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 $^{113}\text{In}(n,n')^{113m}\text{In}$ and $^{115}\text{In}(n,n')^{115m}\text{In}$ 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 $^{64}\text{Zn}(n,p)^{64}\text{Cu}$ cross section for $2.76^{+0.01}_{-0.02}$ 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 $^{47}\text{Ti}(n,p)^{47}\text{Sc}$ 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.

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1. 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 g/cm² as compared to 10's of mg/cm²), together with the fact that isotope production facilities often produce large secondary neutron fields. Particular interest has been paid to (n,p) and (n,a) 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 $D(T,n)\alpha$ ("DT") and $D(D,n)^3He$ ("DD") reactions. While reactors produce copious quantities of neutrons, their energy spec-

E-mail address: andrew.voyles@berkeley.edu (A.S. Voyles).

tra 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 [1]. 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 $(n,\alpha xn)$ 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

^{*} Corresponding author.



Shamsuzzoha Basunia <sbasunia@lbl.gov>
To: Andrew Voyles <andrew.voyles@gmail.com>

Fri, Jun 15, 2018 at 2:39 PM

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Ph.D. Candidate, Nuclear Engineering
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3115B Etcheverry Hall
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850-281-0217 | andrew.voyles@gmail.com

--

M. Shamsuzzoha Basunia Nuclear Science Division, MS 88RO192 One Cyclotron Road Lawrence Berkeley National Laboratory Berkeley, California 94720-8101 USA

Tel: 510 486 7648 FAX: 510 486 7983



Jon Charles Batchelder <batchelder@berkeley.edu>

Wed, Jun 20, 2018 at 2:29 PM

To: Andrew Voyles <andrew.voyles@gmail.com>, Andrew Voyles <ANDREW.VOYLES@berkeley.edu>

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Bauer, Joseph D. <bauer1@llnl.gov> To: Andrew Voyles <andrew.voyles@gmail.com> Cc: Lee Bernstein <labernstein@berkeley.edu>, Karl van Bibber <karl.van.bibber@be< th=""><th>Fri, Jun 15, 2018 at 4:22 PM rkeley.edu></th></karl.van.bibber@be<></labernstein@berkeley.edu></andrew.voyles@gmail.com></bauer1@llnl.gov>	Fri, Jun 15, 2018 at 4:22 PM rkeley.edu>
Andrew,	
That is fine by me.	
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Joe	
From: Andrew Voyles <andrew.voyles@gmail.com> Sent: Friday, June 15, 2018 1:46 PM To: Shamsuzzoha Basunia <sbasunia@lbl.gov>; Jon Charles Batchelder <batchelde <baseling="" a="" be="" common="" of="" of<="" state="" td="" the="" to=""><td>bl.gov>; Eric Matthews I.rutte@berkeley.edu>; MAURICIO bberkeley.edu></td></batchelde></sbasunia@lbl.gov></andrew.voyles@gmail.com>	bl.gov>; Eric Matthews I.rutte@berkeley.edu>; MAURICIO bberkeley.edu>
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 $850\mbox{-}281\mbox{-}0217$ | andrew.voyles@gmail.com



Tim Becker <tbecker@bgc.org>
To: Andrew Voyles <andrew.voyles@gmail.com>

Sat, Jun 16, 2018 at 8:59 AM

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Fri, Jun 15, 2018 at 3:34 PM

To: Andrew Voyles <andrew.voyles@gmail.com>

Cc: "M. Basunia" <sbasunia@lbl.gov>, Batch <batchelder@berkeley.edu>, "Bauer, Joseph D." <baseline

Becker <tbecker@bgc.org>, Eric Matthews <ericfrank94@berkeley.edu>, Paul Renne prenne@bgc.org>, Daniel Rutte

<daniel.rutte@berkeley.edu>, Mauricio Ayllon Unzueta <mayllon@berkeley.edu>, Karl Van Bibber

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- cheers,

Lee Bernstein LBNL Nuclear Data Group Leader Office: +1-(510)-486-4951

Adjunct Professor UC Berkeley Dept. of Nuclear Engineering Office: +1-(510)-642-5107



Eric Matthews <ericfrank94@berkeley.edu>
To: Andrew Voyles <andrew.voyles@gmail.com>

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__

Eric Matthews
UC Berkeley - Nuclear Engineering
ericfrank94@berkeley.edu
510-931-3062



Paul Renne prenne@bgc.org>

Fri, Jun 15, 2018 at 8:33 PM

To: Andrew Voyles <andrew.voyles@gmail.com>

Cc: Shamsuzzoha Basunia <sbasunia@lbl.gov>, Jon Charles Batchelder <batchelder@berkeley.edu>, "Bauer, Joseph D." <baseling

chauer1@llnl.gov>, Tim Becker

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daniel.rutte@berkeley.edu>, MAURICIO AYLLON UNZUETA

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Daniel Rutte <drutte@bgc.org> To: Andrew Voyles <andrew.voyles@gmail.com> Fri, Jun 15, 2018 at 10:32 PM

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Mauricio Ayllon Unzueta <mayllon@berkeley.edu> To: Andrew Voyles <andrew.voyles@gmail.com> Sat, Jun 16, 2018 at 4:08 AM

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On Fri, Jun 15, 2018, 11:46 PM Andrew Voyles <andrew.voyles@gmail.com> wrote: | Hi all,

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Andrew S. Voyles



Karl van Bibber <karl.van.bibber@nuc.berkeley.edu>
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Karl A. van Bibber

Associate Dean for Research, College of Engineering
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Christina Castellanos, Chair's Assistant Department of Nuclear Engineering, University of California, Berkeley 4153 Etcheverry Hall, Berkeley, CA 94720-1730

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Lee Bernstein labernstein@lbl.gov

Fri, Jun 15, 2018 at 3:36 PM

To: Andrew Voyles <andrew.voyles@gmail.com>

Cc: "Birnbaum, Eva R" <eva@lanl.gov>, Jonathan Engle <jwengle@wisc.edu>, stephenagraves <stephenagraves@gmail.com>, Toshihiko Kawano <kawano@lanl.gov>, Amanda Lewis <amanda_lewis@berkeley.edu>, Meiring Nortier <meiring@lanl.gov>

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- cheers,

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Adjunct Professor UC Berkeley Dept. of Nuclear Engineering

Office: +1-(510)-642-5107



Birnbaum, **Eva R** <eva@lanl.gov>
To: Andrew Voyles <andrew.voyles@gmail.com>

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Best.

Eva

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Eva Birnbaum, PhD Program Manager, Isotope Production and Distribution Program Science Program Office Los Alamos National Laboratory MS J975 Phone: 505-665-7167

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If foreign correspondence: Correspondence/TSPA

From: Andrew Voyles <andrew.voyles@gmail.com>

Date: Friday, June 15, 2018 at 2:46 PM

To: Lee Bernstein logov, Eva Birnbaum <eva@lanl.gov, Jonathan Engle jwengle@wisc.edu, Stephen Graves logov, "Kawano, Toshihiko" kawano@lanl.gov, Amanda Lewis logov, Meiring Nortier meiring@lanl.gov

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Jonathan Engle <jwengle@wisc.edu>

Fri, Jun 15, 2018 at 7:36 PM

To: Lee Bernstein < labernstein@lbl.gov>

Cc: Amanda Lewis <amanda_lewis@berkeley.edu>, Andrew Voyles <andrew.voyles@gmail.com>, "Birnbaum, Eva R" <eva@lanl.gov>, Jonathan Engle <jwengle@wisc.edu>, Meiring Nortier <meiring@lanl.gov>, Toshihiko Kawano <kawano@lanl.gov>, stephenagraves <stephenagraves@gmail.com>

Andrew.

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Jon

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The University of California, Berkeley
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- cheers,

Lee Bernstein LBNL Nuclear Data Group Leader Office: +1-(510)-486-4951

Adjunct Professor UC Berkeley Dept. of Nuclear Engineering

Office: +1-(510)-642-5107



Stephen Graves <stephenagraves@gmail.com>
To: Andrew Voyles <andrew.voyles@gmail.com>

Fri, Jun 15, 2018 at 2:26 PM

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All the best,

Andrew S. Voyles



Kawano, **Toshihiko** <kawano@lanl.gov>
To: Andrew Voyles <andrew.voyles@gmail.com>

Sun, Jun 17, 2018 at 11:04 PM

I agree. Thank you,

T. Kawano

From: Andrew Voyles <andrew.voyles@gmail.com>

Date: Friday, Jun 15, 2018, 22:46

To: Lee Bernstein lee Bernstein@lbl.gov, Birnbaum, Eva R <eva@lanl.gov, Jonathan Engle jwengle@wisc.edu, Stephen Graves stephenagraves@gmail.com, Kawano, Toshihiko kawano@lanl.gov, Amanda Lewis amanda_lewis@berkeley.edu, Nortier, Meiring meiring@lanl.gov

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Andrew S. Voyles



Amanda Lewis <amanda_lewis@berkeley.edu>
To: Andrew Voyles <andrew.voyles@gmail.com>

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On Fri, Jun 15, 2018 at 2:46 PM Andrew Voyles <andrew.voyles@gmail.com> wrote:

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I'm planning on including material from our recent Nb(p,x) manuscript as part of my dissertation. To do so, I need to get statements granting me permission to use and reproduce the material as part of my dissertation from all co-authors. Emails from co-authors giving permission will be accepted.

If you're okay with that, can you please respond back with a short statement agreeing to this release? Thanks!!!

All the best,

Andrew S. Voyles



Nortier, Meiring <meiring@lanl.gov>

Mon, Jun 18, 2018 at 5:35 AM

To: Lee Bernstein labernstein@lbl.gov, Andrew Voyles andrew.voyles@gmail.com Co: "Birnbaum, Eva R" <eva@lanl.gov, Jonathan Engle <jwengle@wisc.edu, stephenagraves stephenagraves@gmail.com, "Kawano, Toshihiko" kawano@lanl.gov, Amanda Lewis kawano@lanl.gov, Amanda Lewis kawano@lanl.gov, Amanda Lewis kawano@lanl.gov, Amanda Lewis kawano@lanl.gov, Amanda Lewis kawano@lanl.gov), Amanda Lewis kawano@lanl.gov)

Andrew,

You have my consent to reproduce the data as part of your dissertation.

Best, Meiring

Sent with BlackBerry Work (www.blackberry.com)

From: Lee Bernstein labernstein@lbl.gov

Date: Friday, Jun 15, 2018, 4:37 PM

To: Andrew Voyles <andrew.voyles@gmail.com>

Cc: Birnbaum, Eva R <eva@lanl.gov>, Jonathan Engle <jwengle@wisc.edu>, stephenagraves <stephenagraves@gmail.com>, Kawano, Toshihiko <kawano@lanl.gov>, Amanda Lewis

<amanda lewis@berkeley.edu>, Nortier, Meiring <meiring@lanl.gov>

Subject: Re: Permission to Include Previously Published Material: 10.1016/j.nimb.2018.05.028

I consent to the reproduction of this data for your dissertation!

On Fri, Jun 15, 2018 at 1:46 PM Andrew Voyles <andrew.voyles@gmail.com> wrote:

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All the best,

Andrew S. Voyles

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- cheers.

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