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Proton-induced reactions on Fe, Cu, & Ti from threshold to 55 MeV

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Abstract Theoretical models often differ significantly from measured data in their predictions of the magnitude of nuclear reactions that produce radionuclides for medical, research, and national security applications. In this paper, we compare *a priori* predictions from several state-of-the-art reaction modeling packages (CoH, EMPIRE, TALYS, and ALICE) to cross sections measured using the stacked-target activation method. The experiment was performed using the LBNL 88-Inch Cyclotron with beams of 25 and 55 MeV protons on a stack of iron, copper, and titanium foils. 34 excitation functions were measured for $4 < E_p < 55$ MeV, including the first measurement of the independent cross sections for ^{nat}Fe(p,x)^{49,51}Cr, ^{51,52m,52g,56}Mn, and ^{58m,58g}Co. All of the models failed to reproduce the isomer-to-ground state ratio for reaction channels at compound and pre-compound energies, suggesting issues in modeling the deposition or distribution of angular momentum in these residual nuclei.

1 Introduction

Clinical practice of nuclear medicine is rapidly growing with the inclusion of a broader array of radiopharmaceuticals. Future growth is anticipated, given the pre-clinical success of many new and emerging radionuclides. Although the physical and chemical properties of these novel radionuclides tend to be well-established, their broad-scale clinical applications are reliant upon well-characterized nuclear data to facilitate production.

2 Experimental Methods and Materials

The work described herein follows the methods utilized in our recent work and established by Graves *et al.* for monitor reaction characterization of beam energy and fluence in stacked target irradiations [1, 2]. Preliminary results were reported in a Master's thesis [3]; here we report the final analysis of that work. Unless otherwise stated, all values are presented herein as mean ± SD, or as the calculated result ± half the width of a 68% confidence interval.

2.1 Stacked-target design

We constructed a pair of target stacks for this work, one stack covering the 55–20 MeV range and the other 25–0 MeV. This minimized the systematic uncertainties associated with significant degradation of beam energy, and included

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multiple overlapping measurements between 20–25 MeV as a consistency check between the stacks. A series of nominal 25 µm ^{nat}Fe foils (99.5%, lot #LS470411), 25 µm ^{nat}Ti foils (99.6%, lot #LS471698), and 25 µm ^{nat}Cu foils (99.95%, lot #LS471698) were used (all from Goodfellow Corporation, Coraopolis, PA 15108, USA) as targets. In each stack, seven foils were cut down to 2.5×2.5 cm squares and spatially characterized at four different locations using a digital caliper and micrometer (Mitutoyo America Corp.). Four mass measurements were performed using an analytical balance in order to determine their areal density. The foils were sealed into "packets" using two pieces of 3M 5413-Series Kapton polyimide film tape consisting 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 1.5875 mm-thick aluminum frames. Plates of 6061 aluminum alloy served as proton energy degraders between energy positions. The target box, seen in **??**, is machined from 6061 aluminum alloy and mounts on the end of an electrically-isolated beamline. The specifications of both target stacks are in Table 2 of **??**.

2.2 Quantification of induced activities

A single ORTEC GMX Series (model #GMX-50220-S) High-Purity Germanium (HPGe) detector was used to determine the activities in each target. Samples were counted at fixed positions ranging 5–60 cm (5% maximum permissible dead-time) from the front face of the detector. The foils were counted for 4 weeks following (EoB). An example of one of the gamma-ray spectra collected is shown in **??**. Net peak areas were fitted using the code FitzPeaks [4], which utilizes the SAMPO fitting algorithms for gamma-ray spectra [5].

2.3 Proton fluence determination

Thin ^{nat}Ti and ^{nat}Cu foils were co-irradiated to measure beam current at each position within the stack. The IAEA-recommended $^{nat}Ti(p,x)^{46}Sc$, $^{nat}Ti(p,x)^{48}V$, $^{nat}Cu(p,x)^{62}Zn$, and $^{nat}Cu(p,x)^{63}Zn$ monitor reactions were used [6]. Systematically enhanced fluence from $^{nat}Ti(p,x)^{48}Sc$ co-production was avoided by only using the 928.327, 944.130, and 2240.396 keV decay gammas from ^{48}V. Using the formalism outlined in our previous work, the integral form of the well-known activation equation was used to determine proton fluence ($I\varDelta t$), in order to account for energy loss across each monitor foil [1]. The propagated uncertainty in proton fluence is calculated as the quadrature sum of (1) the uncertainty in quantified EoB activity, (2) uncertainty in the duration of irradiation (conservatively estimated at 10 s, to account for any transient changes in beam current), (3) uncertainty in foil areal density, (4) uncertainty in monitor product half-life (included, but normally negligible), (5) uncertainty in IAEA recommended cross section (using values from the 2017 IAEA re-evaluation [6]), and (6) uncertainty in differential proton fluence (from transport simulations).

2.4 Proton transport calculations

Estimates of the proton beam energy for preliminary stack designs were calculated using the Anderson & Ziegler (A&Z) stopping power formalism [7–9]. However, the transport code FLUKA-2011.2x.3 was used for simulation of the full 3-D target stack and to determine the full proton energy and fluence distribution for each foil [10]. 10^8 source protons were used for all FLUKA simulations, yielding a statistical uncertainty of less than 0.01%. As with the determination of proton fluence in the monitor foils, the progressively increasing energy straggle towards the rear of each stack is accounted for using FLUKA. These energy distributions $\frac{d\phi}{dE}$ were used to calculate a flux-weighted average proton energy $\langle E \rangle$, which accounts for the slowing-down of protons within a foil (particularly in the low-energy stack) and reports the effective energy centroid for each foil. To report a complete description of the representative energy for each foil, a bin width is provided through the energy uncertainty, calculated as the full width at half maximum (FWHM) of the FLUKA-modeled energy distribution for each foil.

2.5 Calculation of measured cross sections

Using the quantified EoB activities along with the variance-minimized proton fluence, it is possible to calculate cross sections for observed (p,x) reactions. While thin ($\approx$ 10–20 mg/cm^2) foils were irradiated to minimize the energy bins of these cross section measurements, all cross sections reported here are flux-averaged over the energy distribution subtended by each foil. The beam current, measured using a current integrator connected to the electrically-isolated target box, remained stable for the duration of the 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 (conservatively estimated at 10 s, to account for any minor 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).

3 Results and Discussion

3.1 Measurement of nuclear excitation functions

After irradiation, all foils were still sealed in their Kapton packets, verifying that no activation products were lost due to packet failure. With the exception of a single foil (Cu-20, in the 25 MeV stack), each activated foil had a small "blister" under the Kapton tape layer, caused by a combination of off-gassing of oxides and the formation of gaseous short-lived beta activities in the tape. This blister verifies that the primary proton beam was incident upon the foil, and provides additional evidence that the beam was stopped in the stack between Ti-20 and Cu-20. Using the ^{nat}Ti(p,x)^{46}Sc, ^{nat}Ti(p,x)^{48}V, ^{nat}Cu(p,x)^{62}Zn, and ^{nat}Cu(p,x)^{63}Zn monitor reactions, as discussed in **??**, a fluence of 17.9 ± 1.0 nAh was calculated to be incident upon the 55 MeV target stack using the FLUKA fluence model, and a fluence of 19.0 ± 1.3 nAh using the linear fit model. Similarly, for the 25 MeV stack, a fluence of 27.5 ± 8.3 nAh was calculated to be incident upon the target stack using the FLUKA fluence model, and a fluence of 31.7 ± 3.7 nAh using the linear fit model to the four frontmost compartments (before the fluence loss becomes strongly nonlinear). Both linear models are consistent with the nominal fluence of 20.78 nAh (for the 55 MeV stack) and 31.61 nAh (for the 25 MeV stack) measured using the current integrators. However, for both target stacks, the FLUKA transport model predicts a significant increase in proton fluence, in particular for the 25 MeV stack. This model fails to reproduce the fluence loss seen in monitor foils, and predicts a significantly higher production of lower-energy secondary protons not seen in the activation data. As fluence loss scales with $\sigma_{\text{tot}}\rho\Delta r$, it is expected that an extrapolation back to the stack entrance (through the SS-3/SS-5 profile monitors) will underestimate the nominal fluence incident upon the box. This incident fluence dropped by approximately 8.9% to 17.3 ± 1.5 nAh using the linear fit model over the length of the 55 MeV stack, which is consistent with similar measurements at the Los Alamos National Laboratory's Isotope Production Facility in the past [1, 2]. 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.

3.2 Comparison of reaction modeling with experimental results

The measured cross sections were compared to the predictions by the reaction codes TALYS, EMPIRE, CoH, ALICE, and by the calculations in the TENDL database. The codes were all run on their default settings, in order to assess their predictive capabilities for the casual user. The default settings for the optical models and gamma strength function (γSF) are listed in Table 1. The level density models for each are as follows. For both CoH and TALYS, the default level density model is the Gilbert-Cameron (GC) model [23], which uses the Constant Temperature model at lower excitation energies and the Fermi Gas model at higher energies. In EMPIRE, the

Table 1: Default settings for the reactions codes

Code Version	Proton/Neutron Optical Model	Alpha Optical Model	E1 γSF Model
EMPIRE-3.2.3[11]	Koning-Delaroche[12]	Avrigeanu(2009)[13]	Modified Lorentzian[14]
TALYS-1.8[15]	Koning-Delaroche	Specific folded potential[15]	Brink-Axel Lorentzian[15]
CoH-3.5.3[16, 17]	Koning-Delaroche	Avrigeanu(1994)[18]	Generalized Lorentzian[16, 17]
ALICE-2017[19]	Nadasen[20]	Parabolic Diffuse-Well[21]	Berman-Fultz Lorentzian[22]

default level density model is the Enhanced Generalized Superfluid Model (EGSM) [24]. This model uses the Generalized Superfluid Model (GSM) [25, 26] at lower energies and the Fermi Gas model as well at higher energies, and has been normalized to discrete levels. This normalization is performed in such a way that it only affects the level density below the neutron separation energy. Finally, the default level density model in ALICE is the Kataria-Rarnamurthy-Kapoor (KRK) model [27, 28], a semi-empirical nuclear level density formula which provides shell-dependent corrections to the nuclear mass surface, based on a Fourier expansion of the single particle level density of nucleons.

4 Conclusions

We present here a set of measurements of 34 cross sections for the $^{\mathrm{nat}}$Fe(p,x), $^{\mathrm{nat}}$Cu(p,x), and $^{\mathrm{nat}}$Ti(p,x) reactions up to 55 MeV, as well as independent measurements of three isomer branching ratios. Nearly all cross sections have been reported with higher precision than previous measurements. We report the first measurements for $\leq$70 MeV protons of the $^{\mathrm{nat}}$Fe(p,x)^{49}Cr, $^{\mathrm{nat}}$Fe(p,x)^{51}Mn, $^{\mathrm{nat}}$Fe(p,x)$^{52\mathrm{m}}$Mn, $^{\mathrm{nat}}$Fe(p,x)^{56}Mn, and $^{\mathrm{nat}}$Fe(p,x)$^{58\mathrm{m}}$Co reactions, as well as the first measurement of the independent cross sections for $^{\mathrm{nat}}$Fe(p,x)^{51}Cr, $^{\mathrm{nat}}$Fe(p,x)$^{52\mathrm{g}}$Mn, $^{\mathrm{nat}}$Fe(p,x)$^{58\mathrm{g}}$Co, and the $^{52\mathrm{m}}$Mn (2^+) / $^{52\mathrm{g}}$Mn (6^+) and $^{58\mathrm{m}}$Co (5^+) / $^{58\mathrm{g}}$Co (2^+) isomer branching ratios via $^{\mathrm{nat}}$Fe(p,x). We also use these measurements to illustrate the deficiencies in the current state of reaction modeling up to 55 MeV for $^{\mathrm{nat}}$Fe(p,x), $^{\mathrm{nat}}$Cu(p,x), and $^{\mathrm{nat}}$Ti(p,x) reactions. Finally, this work provides another example of the current issues with modeling of stopping power in stacked target charged particle irradiation experiments, corrected using variance minimization techniques.

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Data availability. The gamma-ray spectra and all other raw data created during this research are openly available at: `XXXXX` []. All other derived data are available from the corresponding author on reasonable request.

See Supplemental Material at [URL will be inserted by publisher] for a tabulation of the relevant nuclear data used in the analysis for the present work.

Appendix A: Stack design

Table 2: Specifications of the 25 MeV and 55 MeV target stack designs in the present work. The proton beam enters the stack upstream of the SS-5 and SS-3 profile monitors, respectively, and travels through the stack in the order presented here. The 6061 aluminum degraders have a measured density of approximately 2.68 ± 0.03 g/cm^3. Their areal densities were determined using the variance minimization techniques described in this work and an earlier paper [1]. A 316 stainless steel foil is inserted at both the front and rear of each target stack as a monitor of the beam's spatial profile, by developing radiochromic film (Gafchromic EBT3) after end-of-bombardment (EoB).

25 MeV Target layer	Measured thickness	Measured areal density (mg/cm^2)	Uncertainty in areal density (%)	55 MeV Target layer	Measured thickness	Measured areal density (mg/cm^2)	Uncertainty in areal density (%)
SS profile monitor SS-5	130.94 µm	100.57	0.17	SS profile monitor SS-3	130.9 µm	100.48	0.17
Fe-08	26.25 µm	19.69	0.17	Fe-01	25.75 µm	20.22	0.21
Ti-14	25.01 µm	10.87	0.36	Ti-01	25.88 µm	11.09	0.16
Cu-14	24.01 µm	17.49	0.40	Cu-01	28.81 µm	22.40	0.11
Al Degrader E-09	256.5 µm	–	–	Al Degrader A-1	2.24 mm	–	–
Fe-09	26.5 µm	19.90	0.09	Fe-02	25.5 µm	19.91	0.13
Ti-15	23.81 µm	10.97	0.11	Ti-02	25.74 µm	10.94	0.24
Cu-15	21.81 µm	17.63	0.46	Cu-02	28.75 µm	22.32	0.40
Al Degrader H-01	127.09 µm	–	–	Al Degrader A-2	2.24 mm	–	–
Fe-10	26.5 µm	19.84	0.11	Fe-03	25.25 µm	20.00	0.27
Ti-16	24.6 µm	10.96	0.32	Ti-03	25.91 µm	11.25	0.15
Cu-16	22.01 µm	17.22	0.25	Cu-03	28.86 µm	22.49	0.20
Fe-11	27.26 µm	19.96	0.17	Al Degrader C-1	0.97 mm	–	–
Ti-17	25.01 µm	10.88	0.25	Fe-04	25.25 µm	19.93	0.33
Cu-17	29 µm	21.91	0.33	Ti-04	25.84 µm	10.91	0.18
Fe-12	27.01 µm	20.03	0.12	Cu-04	28.78 µm	22.38	0.29
Ti-18	25.01 µm	11.00	0.87	Al Degrader C-2	0.97 mm	–	–
Cu-18	28.75 µm	22.33	0.14	Fe-05	25.64 µm	20.02	0.24
Fe-13	26.25 µm	20.05	0.16	Ti-05	25.86 µm	10.99	0.30
Ti-19	26.6 µm	11.01	0.22	Cu-05	28.77 µm	22.35	0.12
Cu-19	28.75 µm	22.32	0.19	Al Degrader C-3	0.97 mm	–	–
Fe-14	25.75 µm	20.11	0.19	Fe-06	25.75 µm	20.21	0.26
Ti-20	27.01 µm	11.06	0.35	Ti-06	25.5 µm	11.15	0.23
Cu-20	28.26 µm	22.34	0.28	Cu-06	28.83 µm	22.43	0.10
SS profile monitor SS-6	131.5 µm	100.99	0.17	Al Degrader C-4	0.97 mm	–	–
				Fe-07	25.76 µm	19.93	0.19
				Ti-07	25.75 µm	11.17	0.33
				Cu-07	28.76 µm	22.34	0.24
				Al Degrader H-02	127.04 µm	–	–
				SS profile monitor SS-4	131.21 µm	101.25	0.16

Appendix B: Measured excitation functions

Figures of the cross sections measured in this work are presented here, in comparison with literature data

Appendix C: 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 [29–32].

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