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

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**[sec:intro\_fe]Introduction**

The next generation of nuclear medicine is likely to involve the production of novel, emerging radionuclides . These make possible an improvement in clinical practice, offering many patients the possibility for better survival rates and quality lives, and informing researchers in their quest to develop new treatments . While the chemical and radioactive decay properties of many of these radionuclides are well-established, their broad-scale clinical applications are reliant upon well-characterized nuclear data to facilitate large-scale production. One particular group of emerging radionuclides are positron-emitting isotopes of manganese, which have been identified as having potential for a range of diagnostic applications . In particular, a significant interest has been expressed in producing the emerging radionuclides for clinical use in metabolic PET studies, as well as for pre-clinical imaging of neural and immune processes via PET .

However, the modeling of nuclear reactions in this mass region presents numerous challenges including uncertainties in nuclear level densities as a function of spin due to the opening of the *f*7/2 orbital and the presence of enhanced *γ*-strength at low energies . Therefore, as part of a larger campaign to address deficiencies in cross-cutting nuclear data needs, our group has performed a measurement of the nuclear excitation functions of the radionuclides , , and . This was carried out using the thin-foil stacked-target technique to study proton-induced reactions on natural iron foils with titanium and copper monitor foils. This work seeks to complement earlier measurements at 40–100 MeV proton beam energies and extend them down to reaction thresholds, in order to investigate the feasibility of production using the international network of low-energy medical cyclotrons . Furthermore, we are able to use a portion of the data we measured to gain insight into the role of angular momentum in the transitional energy region between compound and direct reactions since both the ground and long-lived isomeric states in were populated.

Manganese radionuclides are desirable for radiopharmaceutical applications, as the element is chemically versatile, biologically relevant, and essential in trace quantities. Manganese possesses well-established biochemistry, and has been proven to be chelated well by DOTA for tracking monoclonal antibodies, with high biostability at neutral pH . (*t*1/2 = 5.591±0.003 d, *Iβ*+ = 29.4%, *Eβ*, avg = 0.242 MeV) has been shown to be useful for immunoPET applications, with its rapid blood clearance offering the possibility for imaging within minutes of injection, making it more useful for imaging than the more established immunoPET agents or , and highly suitable for pre-clinical imaging . Furthermore, has high uptake in the heart, liver, kidneys, and pancreas, making it a useful diagnostic agent for pancreatic *β*-cell, insulinoma, and porphysome imaging . However, its long lifetime and unfavorable high-energy decay gamma-rays make it undesirable for clinical applications. The short half-life of the isomer (*t*1/2 = 21.1±0.2 min) makes production and handling difficult, and its high-intensity gamma emission (1434.06 keV [*Iγ* = 98.2 ± 0.5%]) contributes significantly to patient dose . In contrast, (*t*1/2 = 46.2±0.1 min, *Iβ*+ = 97.09%, *Eβ*, avg = 0.973 MeV), is more clinically suitable for rapid metabolic studies . lacks any strong decay gamma-rays (its longer-lived daughter [*t*1/2 = 27.704±0.003 d] has only a single 320.0284 keV [*Iγ* = 9.910 ± 0.010%] line), making it the best choice of these radionuclides for clinical imaging,.

In addition to their interest for PET studies, the excitation functions offer an opportunity to study the distribution of angular momentum in compound nuclear and direct pre-equilibrium reactions via observation of the (*t*1/2 = 21.1±0.2 min; J*π* = 2+) to (*t*1/2 = 5.591±0.003 d; J*π* = 6+) ratio . 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 . These measurements also provide an opportunity to benchmark the predictive capabilities of reaction modeling codes used for nuclear reaction evaluations and the way in which they implement the underlying physical reaction mechanisms.

**[sec:experimental\_fe]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 . Preliminary analysis was previously reported in the Master’s thesis of one co-author, but the conclusive results of this work are described here .

**[sec:target\_design\_fe]Stacked-target design**

A pair of target stacks were constructed for this work. One stack covered the 55–20 MeV range and the other 25–0 MeV, in order to minimize the systematic uncertainties associated with significant degradation of beam energy, and includes multiple overlapping measurements between 20–25 MeV to build confidence in the results. A series of nominal 25 m foils (99.5%, lot #LS470411), 25 m foils (99.6%, lot #LS471698), and 25 m 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 m of a silicone adhesive (nominal 4.79 mg/cm2) on 25.4 m of a polyimide backing (nominal 3.61 mg/cm2). 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 stack designs for this work are presented in of Appendix [8](#sec:fe_stack_design).

Both target stacks were separately irradiated at the Lawrence Berkeley National Laboratory (LBNL) 88-Inch Cyclotron, a K=140 sector-focused cyclotron . The 25 MeV stack was irradiated for approximately 20 minutes at a nominal current of 100 nA, for an integral current of 31.61 nAh, measured using a current integrator on the electrically-isolated beamline. The 55 MeV stack was irradiated for approximately 10 minutes at a nominal current of 120 nA, for an integral current of 20.78 nAh. The beam current remained stable under these conditions for the duration of each irradiation. The approximately 1 cm-diameter proton beam incident upon each stack’s upstream stainless steel profile monitor had a maximum energy of either 25 or 55 MeV, with an approximately 2% energy width due to multi-turn extraction — these energy profiles were used for all later analysis. Following end-of-bombardment (EoB), each stack was removed from the beamline and disassembled. All activated foils were transported to a counting lab for gamma spectrometry, which started approximately 30 minutes following the end of each irradiation.

**[sec:spectroscopy\_fe]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, with a series of standard calibration sources used to determine energy and efficiency for each position. The foils were counted for 4 weeks following end-of-bombardment (EoB). An example of one of the gamma-ray spectra collected is shown in . Net peak areas were fitted using the gamma spectrometry analysis code FitzPeaks , which utilizes the SAMPO fitting algorithms for gamma-ray spectra .

[image: ./figures/sample\_gspec\_fe.pdf]

[fig:gspec\_femn]

The net counts in each fitted gamma-ray photopeak were converted into activities for the decaying activation products. The lifetimes and gamma-ray branching ratios used for all calculations of measured cross sections reported in this work have been taken from the most recent edition of Nuclear Data Sheets for each mass chain . Corrections for gamma-ray attenuation within each foil packet were made, using photon attenuation coefficients from the XCOM photon cross sections database . The total propagated uncertainty in 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.

As in our previous work, these activities were used to calculate cumulative and independent cross sections . For the first observable product nuclide in a mass chain, its (p,x) cross section is reported as a cumulative cross section (*σc*), 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 are 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 (*σi*) corresponding to a single exit channel are reported, facilitating comparison to reaction model calculations. Solutions to the first- and higher-order Bateman equations are used for separation of feeding contributions from decay precursors, so that independent cross sections may be reported .

**[sec:dosimetry\_fe]Proton fluence determination**

In addition to measuring beam current using a current integrator, thin and foils were included along with the targets, to monitor beam current at each position within the stack. The IAEA-recommended (p,x), (p,x), (p,x), and (p,x) monitor reactions were used . Using the formalism outlined in our previous work, the integral form of the well-known activation equation was used to determine proton fluence (*IΔt*), in order to account for energy loss across each monitor foil . 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 ), and (6) uncertainty in differential proton fluence (from transport simulations).

**[sec:proton\_transport\_fe]Proton transport calculations**

Estimates of the proton beam energy for preliminary stack designs were calculated using the Anderson & Ziegler (A&Z) stopping power formalism . 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 . 108 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 ⟨*E*⟩, 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.

The “variance minimization” techniques utilized in our recent work and established by Graves *et al.* have been used to reduce uncertainty in proton fluence assignments . This method is based on the assumption that the independent measurements of proton fluence from the different monitor reactions should all be consistent at each position. Any residual disagreement in the observed proton fluences is primarily due to poorly-characterized stopping power in simulations or a systematic error in the areal densities of the stack components. This disagreement is minor at the front of the stack, but gets progressively worse as the beam travels through the stack, due to the compounded effect of systematic uncertainties in stack areal densities.

When performing a variance minimization, it is important to apply this variation of effective areal density to the stack components which have the most significant impact on beam energy loss. Therefore, the aluminum degraders are used for variance minimization for the 55 MeV stack, as they make up more than 80% of the areal density of the stack. For the 25 MeV stack, the Kapton tape was chosen for variance minimization, as the foil packets themselves are responsible for the majority of beam degradation. While it only makes up approximately 20% of the low-energy stack’s areal density, the Kapton surrounding each foil packet has a greater areal density than the foil itself. In addition, it is far easier to directly characterize the areal density of the metallic foils than it is for the Kapton, resulting in only an approximate value for the latter. While it might seem obvious, the contributions to the slowing of the beam due to the adhesive has often been neglected in much work performed to date. This is of relatively minor consequence for higher-energy irradiations (especially relative to any beam degraders), but becomes increasingly important for proton energies below approximately 25 MeV, causing as much as a loss of 8 MeV by the rear of this stack, making the precise areal density a source of significant uncertainty.

In performing the minimization, the areal density of each of the aluminum degraders (for the 55 MeV stack) were varied uniformly in FLUKA simulations by a factor of up to ±25% of nominal values, to find the effective density which minimized variance in the measured proton fluence at the lowest energy position (Ti-07, Cu-07). For the 25 MeV stack, the areal density reached in the minimization of the 55 MeV stack was used for the E-09 and H-01 aluminum degraders and the areal density of each of the Kapton tape layers were varied by ±25% , to find the effective density which minimized variance in the measured proton fluence at the next-to-lowest energy position (Ti-19, Cu-19). These positions were chosen as minimization candidates, as they are the most sensitive to systematic uncertainties in stack design. In the 25 MeV stack, activity was not seen in gamma spectrometry for the lowest-energy (Cu-20) monitor foil, implying that the beam was stopped at some point in between Ti-20 and Cu-20. This observation indicates that the true areal densities of the stack components differ from nominally measured values (primarily for the difficult-to-characterize Kapton tape), as transport calculations using nominal areal densities predict that the beam should exit the stack with an energy of approximately 7 MeV. As a result, this position was not used for minimization, with the Ti-19 and Cu-19 position being the lowest-energy reliable monitor foils in the stack. The results of the minimization technique indicate a clear minimum in proton fluence variance for flux-weighted average 22.71 MeV protons entering the last energy position of the 55 MeV stack. This is approximately 2 MeV lower than the nominal FLUKA simulations, and approximately 2 MeV lower than nominal A&Z calculations, both of which used the nominal 2.80 g/cm3 measured density of the aluminum degraders. This energy corresponds to an aluminum areal density of 4.43% greater than nominal measurements, and corrects for other minor systematic uncertainties in stack design, including stack areal densities and incident beam energy. Similarly, for the 25 MeV stack, variance minimization converges on flux-weighted average 9.23 MeV protons entering the Fe-13/Ti-19/Cu-19 energy position, which is approximately 4 MeV lower than the nominal FLUKA simulations, and approximately 5 MeV lower than nominal A&Z calculations. This energy corresponds to a Kapton tape areal density of 5.69% greater than nominal measurements, which is completely reasonable given the lack of areal density data from the manufacturer. The impact of this variance minimization for improving disagreement in proton fluence is clearly seen in .

[fig:fe\_variance\_mins]

An enhanced version of the final (p,x), (p,x), (p,x), and (p,x) monitor reaction fluences is shown in . The uncertainty-weighted mean for the two (p,x) and two (p,x) monitor channels was calculated at each energy position, to determine the final fluence assignments for the Cu and Ti foils, respectively, and the uncertainty-weighted mean for all four monitor channels was used to determine the final fluence assignments for the Fe foils. Uncertainty in each final proton fluence is calculated by error propagation of the individual monitor channel fluence values at each energy position. These weighted-mean fluences are plotted in , along with the estimated fluence according to both FLUKA 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 for the 55 MeV stack, with the FLUKA model predicting a slightly greater fluence loss throughout the stack. However, neither model is capable of accurately modeling the rapid decrease in apparent fluence at the rear of the 25 MeV stack. These models are used purely to provide an extrapolation from the highest-energy position back to the “front” of each stack, to compare with the nominal fluence measured by the beamline current integrators.

[image: ./figures/fluence\_plot.pdf]

[fig:fe\_fluence\_plot]

**[sec:calcs\_sec\_fe]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 (p,x) reactions. While thin (≈ 10–20 mg/cm2) foils were irradiated to minimize the energy bins 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. For both the cumulative and independent activities quantified, cross sections were calculated using the formalism outlined in our previous work . 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).

**[sec:results\_fe]Results and Discussion**

**Measurement of nuclear excitation functions**

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. 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 thermal swelling and the formation of short-lived beta activities. 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 (p,x), (p,x), (p,x), and (p,x) monitor reactions, as discussed in , a fluence of 18.8±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 33.2±4.7 nAh was calculated to be incident upon the 55 MeV target stack using the FLUKA fluence model, and a fluence of 37.0±9.3 nAh using the linear fit model. All of these 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. As fluence loss scales with *σtotρΔr*, 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 9.5% to 17.0±2.4 nAh (and by 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 . 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 , , , , and were extracted for (p,x) reactions on foils up to 55 MeV, as recorded in . For (p,x) reactions on , the (p,x) cross sections for , , , and were extracted, as recorded in . For (p,x) reactions on , the (p,x) cross sections for and were extracted, as recorded in . 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 (p,x), (p,x), and (p,x) were extracted and are recorded in . Comparisons of the measured cross sections and isomer branching ratios with literature data (retrieved from EXFOR ) are seen in the figures of Appendices [6](#sec:fe_xs_figures) and [7](#sec:fe_ibr_figures). 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 (±2–10%), proton fluence (±5–13%), and foil areal density (±0.1–0.3%).

[tab:fe\_rp\_table]

[tab:cufe\_rp\_table]

[tab:ti\_rp\_table]

[tab:fe\_ibr\_table]

These results have several notable features. The various , , and (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. While (p,x) reactions below 70 MeV on these elements are well-characterized overall, measurements of several reaction channels are somewhat sparse in comparison. Indeed, fewer than four existing measurements have been performed for the (p,x),, (p,x),, and (p,x) reactions presented here. Additionally, activity is seen in all foils, consistent with proton activation of the trace silicon in the Kapton tape used for foil encapsulation, as described in our previous work . No cross sections for (p,x) are reported due to the significant uncertainty in characterizing the layer of silicone adhesive, but this serves as another example of how the use of silicone-based adhesives may systematically enhance the apparent fluence when using the (p,x) monitor reactions.

This work presents the first measurements of several observables in this mass region, including the (p,x), (p,x), (p,x), (p,x), and (p,x) reactions in the 0–70 MeV region, the independent cross sections for (p,x), (p,x), (p,x), and the (2+) / (6+) and (5+) / (2+) isomer branching ratios via (p,x). The cumulative cross sections from these data are also consistent with existing measurements of the cumulative (p,x),, cross sections.

Several of the activities produced via (p,x) could be useful as experimental monitor reactions. In particular, (p,x) (*t*1/2 = 77.236 ± 0.026 d, *ϵ*=100% to  ) is a strongly-fed reaction channel (peak cross section of approximately 400 mb near 12 MeV), with a sufficiently-long lifetime permitting offline gamma spectrometry, and cannot be populated via secondary neutrons incident upon the monitor target. It possesses a number of intense gamma-rays, which are distinct from those populated by daughter states in the decay of , and the gammas which are produced both in the decay of and may be easily resolved based on differences in intensity and lifetime. Similarly, (p,x) (*t*1/2 = 312.20 ± 0.020 d, *ϵ*=100% to  ) has a convenient lifetime and strong cross section (peak of approximately 160 mb near 40 MeV), and is immune from two different reactions on the same monitor foil leading to states in the same daughter nuclide. This reaction could be useful for intermediate- to high-energy protons (*Ep*≥30 MeV), but would be susceptible to production via the high-energy secondary neutrons (threshold 12.1 MeV off of ) produced in these facilities, though (p,x) production rates should dominate (n,x) through both particle flux and cross section.

All: feel free to add additional details of 51/52Mn production here...

Notably, this work is the most well-characterized measurement of the (p,x) reactions below 70 MeV to date, with cross sections measured at the 6–10% uncertainty level. This is important, as it presents the first measurement of the (p,x) reaction, the first measurement of the independent (p,x) cross sections, and extends the (p,x) excitation function down to the lowest energy to date. (p,x) appears to offer a compelling alternative to the more established (d,x) pathway, which necessitates an enriched target to avoid radio-manganese contamination from reactions on stable isotopes of Cr . (p,x) could be used for clean production of (≥98.8% radioisotopic purity) below 20 MeV using the (p,*α*) channel. In addition, this low-energy production is accessible using the international network of small medical and research cyclotrons, enabling in-house production of this short-lived (*t*1/2 = 46.2 ± 0.1 m ) radionuclide. To increase yields over (p,x), an enriched target could be used to take advantage of the eight-fold increase in reaction cross section for production using 40–50 MeV protons, without opening the additional manganese exit channels accessible on a natural target.

Likewise, (p,x) offers an interesting production pathway for . Conventional production uses the low-energy /(p,n) pathways, which offer high radioisotopic purity (approximately 99.6%) . (p,x) offers a nearly threefold increase in production yield, but the low radioisotopic purity (99.1% for 20–30 MeV, decreasing to 60.8% by 40 MeV) at higher energies due to the opening of makes this route seem impractical. Much like , the use of an enriched target would prevent production of , providing a higher-yield production route over (p,x), with the tradeoff of necessitating higher-energy protons (≤35 MeV) for production. It is important to note that the (p,x) route provides ≥60% feeding of (*t*1/2 = 5.591±0.003 d ), implying that the short-lived (*t*1/2 = 21.1±0.2 m ) can be easily separated through the difference in lifetime, to avoid the unfavorable gammas produced by the isomer. However, if nearly pure is desired for preclinical imaging applications, the feeding of through the *ϵ* decay of (*t*1/2 = 8.725±0.008 h ) exclusively populates the isomer, making this potentially suitable for production through “milking” of a generator. Clearly, the use of (p,x) has significant untapped potential, and additional work is needed to further characterize these reaction channels for *Ep*≤60 MeV.

In addition to the (p,x) measurements, this experiment has also yielded measurements of a number of additional emerging radionuclides with medical applications. These include the non-standard positron emitters      ,    , and the *β*−-therapeutic agent  . 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 (p,x), (p,x), and (p,x).

**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 (*γ*SF) are listed in . 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 , which uses the Constant Temperature model at lower excitation energies and the Fermi Gas model at higher energies. In EMPIRE, the default level density model is the Enhanced Generalized Superfluid Model (EGSM) . This model uses the Generalized Superfluid Model (GSM) 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 , 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.

For this analysis, the focus will be on the three largest channels measured – (p,x), (p,x), and (p,x). For the (p,x) reactions, seen in , the lower-energy (p,*α*) reaction, which peaks around 15 MeV, is well-modeled by TALYS and TENDL, and over-predicted by EMPIRE, CoH and ALICE. The higher-energy reactions on the higher mass Fe isotopes, however, are better matched by EMPIRE and CoH. ALICE overpredicts the production of in both peaks, and appears to peak at too high an energy. As was seen in the modeling of high-energy proton-induced niobium reactions , TALYS and TENDL do well with the lower-energy “compound” reactions but do not accurately predict the higher-energy reactions that have a significant pre-equilibrium component. EMPIRE and CoH seem to accurately predict the locations of the peaks, but often fail to reliably estimate the magnitude of the cross section. This is seen again in the (p,x) cross section, where the first peak is well-modeled by TALYS and TENDL, but rises much more rapidly than the data would support above 50 MeV. CoH and EMPIRE, again, overpredict the cross sections at all energies, but seem to have the correct shape. Through measurement of the (p,x) independent cross section, the effect of spin distributions in highly-excited nuclear states can be studied. The measured data suggests that the independent cross section to the 2+ isomer should be a large fraction of the cumulative cross section to (which has a 6+ ground state), over 80% at the peak energy. All three codes predict about 50%, which indicates that the isomer feeding is not well-modeled. It is possible that this is caused by residual nucleus population and/or level density models that are skewed too heavily towards high spin. In the other isomer-to-ground state ratio measured for Fe, (p,x), the opposite is seen – the codes underpredict the ratio of the isomer-to-ground state, but in this case the isomer is the higher spin state (5+, compared with a 2+ ground state). Given that the level density model was the same for all of the product nuclei, it points to the spin distribution of the initial population of the residual nucleus as the main problem with modeling. However, no firm conclusions can be drawn about the direct causes of the inaccuracies, as there are many “moving parts” in these calculations, with measurements performed using natural abundance targets.

[tab:fe\_defaults]

[image: ./figures/51Mn.pdf]

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[fig:temp\_52Mn]

[image: ./figures/52mMn.pdf]

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**[sec:conclusions\_fe]Conclusions**

We present here a set of measurements of 34 cross sections for the (p,x), (p,x), and (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 ≤70 MeV protons of the (p,x), (p,x), (p,x), (p,x), and (p,x) reactions, as well as the first measurement of the independent cross sections for (p,x), (p,x), (p,x), and the (2+) / (6+) and (5+) / (2+) isomer branching ratios via (p,x). We also use these measurements to illustrate the deficiencies in the current state of reaction modeling up to 55 MeV for (p,x), (p,x), and (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.

**Acknowledgements**

The authors would like to particularly acknowledge the assistance and support of Brien Ninemire, Scott Small, Tom Gimpel, and all the rest of the operations, research, and facilities staff of the LBNL 88-Inch Cyclotron. We also wish to acknowledge Haleema Zaneb, who participated in these experiments. This work has been carried out under the auspices of the U.S. Department of Energy by Lawrence Berkeley National Laboratory and the U.S. Nuclear Data Program under contract # DE-AC02-05CH11231. This research was performed under appointment to the Rickover Fellowship Program in Nuclear Engineering, sponsored by the Naval Reactors Division of the U.S. Department of Energy Additional support has been provided by the U.S. Nuclear Regulatory Commission.

This research used the Savio computational cluster resource provided by the Berkeley Research Computing program at the University of California, Berkeley (supported by the UC Berkeley Chancellor, Vice Chancellor for Research, and Chief Information Officer).

**Measured excitation functions**

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

**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 .

**Stack design**

[tab:fe\_stack\_table]

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.