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Deuteron-induced reactions for the production of Neptunium-236m/g

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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

Neptunium-236 is a unique actinide isotope with use in metrology and analytical chemistry applications. Since it is neither anthropogenic nor primordial in occurrence, ^{236}Np constitutes a reference standard for isotope dilution mass spectrometry with the function of improving accuracy and precision for the detection of common neptunium isotopes associated with nuclear energy applications. Isotope dilution is the method of adding a uniquely uncommon isotope of the element whose concentration is to be determined in known amounts to the unknown specimen. Quantification of the relative abundance of the added diluent isotope versus the unknown isotope enables the amount of analyte in the sample to be accurately determined. Isotope dilution mass spectrometry (IDMS) methods, including thermal ionization mass spectrometry (TIMS) and multi-collector inductively-coupled plasma mass spectrometry (MC-ICP-MS), are currently the most sensitive methods for measuring actinides in environmental samples. For quantitative results, an isotope dilution tracer is added to each sample and the ratio of the analyte to the tracer is used to calculate the amount of analyte in a given sample. An ideal IDMS tracer is an isotope that is not found in the sample and is stable or has a long half-life.

However, the current inventory of ^{236g}Np ($t_{1/2} = 1.53\pm0.05 \times 10^5$ y [**?**]) is extremely limited, with the world-wide stockpile limited to 10's of µg of material. The existing U.S. inventory was produced at Argonne National Laboratory in the mid-1980's, via the reaction ^{235}U(d,n)^{236g}Np using a 20 MeV deuteron beam. No effort was made to optimize irradiation conditions to minimize co-production of ^{237}Np or to evaluate alternative production routes. As a result, the current ^{236g}Np stock contains approximately 20% of ^{237}Np (atom %) impurity, which negatively affects both the analytical uncertainty and detection limit for ^{237}Np. Since the small amount of remaining material

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is currently available only for use as an isotope dilution tracer for ^{237}Np determination in high-value samples, it is clear that a new supply of ^{236g}Np tracer is critical to meeting the needs of the applications community. In 2015, the U.S. Certified Reference Materials (CRM) committee's ^{236g}Np working group identified an annual requirement of 10–20 µg / y, with a minimum 93.3 atom% ^{236g}Np content. An exact knowledge of ^{235}U(d,x)^{235,236,237}Np reaction cross sections and threshold energies leads to ^{236g}Np production with maximal yield and isotopic purity. Therefore, as part of a larger campaign to address deficiencies in cross-cutting nuclear data needs, our group has measured the nuclear excitation functions of the radionuclides ^{236}Np from deuteron-induced reactions on highly-enriched ^{235}U (HEU) targets, using a new variant on the stacked-target technique with nickel and titanium monitor foils.

Further, ^{236}Np is an interesting odd-odd nucleus with a high-spin ground state ($J^\pi = (6^-)$ [**?**]), as well as a low-lying isomer resulting from an anti-parallel coupling configuration ($t_{1/2}$ = 22.5±0.4 h, $E_x \approx 70$ keV, $J^\pi = 1$ [**?**]). From an isotope production perspective, ^{236m}Np is interesting, as it has a 50±3% decay branch [**?**] to ^{236}Pu, which is itself a valuable reference tracer for the quantitative assay of Pu isotopes. In addition to its needs for isotope production, this experiment offers an opportunity to study the distribution of angular momentum in compound nuclear and direct pre-equilibrium reactions via observation of the ^{236m}Np to ^{236g}Np 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 [1, 2].

2 Experimental Methods and Materials

The work described herein follows the methods utilized in our recent work for monitor reaction characterization of beam energy and fluence in stacked target irradiations [3**?**]. Unless otherwise stated, all values are presented herein as mean ± SD, or as the calculated result ± half the width of a 1σ confidence interval.

2.1 Stacked-target design

In such a variant, activation foils must by necessity become progressibely smaller in diameter towards the rear of the stack, to ensure that all deuterons incident upon a foil were degraded by all upstream stack elements.

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 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 1 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

Table 1 Specifications of the 16 MeV and 12 MeV "packet" designs in the present work. The "overfilled" deuteron beam enters the stack upstream of the Ni-01 and Ni-02 foils, respectively, and travels through the stack in the order presented here. Note that due to the nonuniform thickness and highly brittle nature of the U-01 and U-02 targets, the listed thicknesses are approximate values obtained through subtraction from the total thickness of the sealed packets. Uncertainties are listed in the least significant digit, that is, 24.6(12) mm means 24.6 ± 1.2 mm.

16 MeV Target layer	Measured diameter (mm)	Measured thickness (mm)	Measured mass (g)	Measured areal density (mg/cm^2)	12 MeV Target layer	Measured diameter (mm)	Measured thickness (mm)	Measured mass (g)	Measured areal density (mg/cm^2)
Ni-01	17.89(23)	0.0273(5)	0.0570(8)	22.68(49)	Ni-02	18.04(30)	0.0267(6)	0.0557(6)	21.78(43)
U-01	12.33(4)	*0.105*	0.2490(2)	208.71(60)	U-02	12.33(4)	*0.084*	0.2010(2)	168.47(48)
Ti-01	11.53(40)	0.0275(6)	0.0113(6)	10.86(67)	Ti-02	10.92(24)	0.0273(6)	0.0101(5)	10.78(58)
Al-01	11.85(30)	0.0283(5)	0.0072(6)	6.53(55)	Al-02	11.78(32)	0.0285(6)	0.0070(6)	6.42(56)

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 ($I\Delta t$), in order to account for energy loss across each monitor foil [3]. 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 $\pm$ 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 $\pm$ 1.3 nAh using the linear fit model. Similarly, for the 25 MeV stack, a fluence of 27.5 $\pm$ 8.3 nAh was calculated to be incident upon the target stack using the FLUKA fluence model, and a fluence of 31.7 $\pm$ 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 $\sigma_{\mathrm{tot}}\rho\Delta r$, 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 $\pm$ 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 [3, 11]. 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.

Mention spin applications for isotope production:

236gNp formation favoured at: increased particle angular momentum input (d,x superior to p,x) Increased particle energy input (m/g ratio decreases with particle energy

The large spin difference between the isomer and ground state has dire

As 236gNp is not populated in any of teh decay modes of 236mNp, it is not possible to simultaneously optimize the production of 236gNp and 236Pu in a single target. However, due to the excitation function of the isomer-to-ground state branching ratio observed in the present work, it would appear possible that a stack of production targets could be used to similar effect, exploiting the enhanced 236gNp production at higher beam energy, and using a lower-energy target downstream where the isomer is more strongly populated.

3.2 236mNp

Comes from gamma spec

3.3 236gNp

Comes from chemistry

3.4 237Np

Comes from chemistry
Mention the subtraction of 237Np from the unirradiated foils
Need for higher-enrichment 235U to produce pure 236Np

3.5 238Np

Comes from chemistry, via 238Pu MS

3.6 Isomer decay branching ratio

Comes from :
total : gamma spec (236mNp)
beta- branch: chemistry (236Pu)

3.7 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 **??**. 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 [12], 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) [13]. This model uses the Generalized Superfluid Model (GSM) [14, 15] 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 [16, 17], 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 ^{nat}Fe(p,x), ^{nat}Cu(p,x), and ^{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 ^{nat}Fe(p,x)^{49}Cr, ^{nat}Fe(p,x)^{51}Mn, ^{nat}Fe(p,x)^{52m}Mn, ^{nat}Fe(p,x)^{56}Mn, and ^{nat}Fe(p,x)^{58m}Co reactions, as well as the first measurement of the independent cross sections for ^{nat}Fe(p,x)^{51}Cr, ^{nat}Fe(p,x)^{52g}Mn, ^{nat}Fe(p,x)^{58g}Co, and the ^{52m}Mn (2^+) / ^{52g}Mn (6^+) and ^{58m}Co (5^+) / ^{58g}Co (2^+) isomer branching ratios via ^{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 ^{nat}Fe(p,x), ^{nat}Cu(p,x), and ^{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: Measured excitation functions

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

Appendix B: 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 [18–21].

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