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Experimental and computational techniques for the analysis of proton beam propagation through a target stack

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ABSTRACT

Proton beam energy, energy straggling, and intensity in thick stacks of target materials at the Los Alamos Isotope Production Facility were investigated using the foil activation technique and computational simulations. Isotopic yield measurements of irradiated foils from several recent experiments used to determine these quantities were compared with the predictions of MCNP6 and TRIM codes, and with Andersen & Ziegler's semi-empirical formalism. Differences between code predictions and experimental data were examined. Methods for computational simulation of energy propagation agree well with one another and were able to accurately predict the proton beam's energy for a limited range. Predictions were accurate when degrading from an initial energy of 100 MeV down to approximately 50 MeV, but struggled to represent measured data well at lower energies.

1. Introduction

Particle transport codes which employ measured or modeled nuclear data to determine the outcome of individual ion-atom probabilistic events are widely used for predicting radiation interaction with materials. One of the more versatile of these codes, Monte Carlo N-Particle (MCNP6) transport code, is based on cross sections for particle interactions [1]. The SRIM/TRIM program suite [2] is based on the parameterization of quantum physical models to experimental data including stopping powers, and is in this way not dissimilar to the semi-empirical formalisms more simply employed by Anderson and Ziegler [3]. As a predictive tool, such calculations are instrumental in the design of radionuclide production targets. These tools are used to calculate energy propagation which aids in cross section measurement experiments. Validation of these calculations against experimental data is therefore critical. We routinely apply MCNP6, TRIM, and A&Z in experiments using the 100 MeV proton beam at the Los Alamos Isotope Production Facility (IPF).

Characterization of the proton energy and intensity as a function of depth in target material is not straight-forward at IPF, where the proton beam is completely stopped by tens of grams per cm^2 target stacks. A stacked-foil activation technique [4] is a convenient method for monitoring the beam energy and intensity at specific locations within a target where Faraday cups and other direct energy measurements may not be possible. In this approach, thin, pure metal foils with well-known production cross sections, acting as targets for nuclear reactions, are irradiated in an experimental stack. By correlating predicted isotopic yields calculated using particle transport codes with yields that are experimentally measured, the effective energy and proton fluence can be determined. Accurate energy and fluence determinations within a specific geometry afford greater certainty for cross section measurements. These cross section measurements can, in turn, be used to optimize target designs for isotope production by exploiting the most productive energy regions of nuclear excitation functions.

Most (p,x) nuclear formation cross sections peak below 100 MeV and their corresponding isotope yields are sensitive to protons with energy in the range of 0–100 MeV. Small variations or straggling in the beam energy incident on a target stack can have drastic effects on the quantity and purity of accelerator produced isotopes. In this work we compared predictions of ^{22}Na production in aluminum monitor foils in a stacked foil experiment with experimentally measured isotope yields. This comparison provided insight into the validity of these computational tools and suggests careful planning is necessary when designing target stacks which attempt to use the entire effective energy range of the IPF proton beam for radioisotope production in thick targets. These considerations are also applicable to other high-energy production facilities including BLIP (US), INR (Russia), ARRONAX (France), TRIUMF (Canada), and iThemba (South Africa).

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In recent years, several proton irradiation experiments [5–9] were conducted at IPF utilizing the stacked foil technique, some with the purpose of measuring cross sections in thin terbium foils [7,10]. Within each of these stacks, aluminum monitor foils were interspersed with the intent of measuring the proton beam intensity at various depths within the target stack. These foils were examined in this work.

In the experiments, the foil stacks were irradiated with a nominal, primary beam energy of 100 MeV, derived from accelerator tuning parameters. Initial predictions of ^{22}Na yields using these beam conditions showed significant discrepancy for downstream aluminum foils at the rear of the target stack. This strongly suggested that the actual beam energy was lower than 100 MeV.

This work established motivation for the implementation of a direct, time-of-flight measurement of the proton beam energy upstream of the target stack. While a time-of-flight measurement improved the agreement for the predicted ^{22}Na production and experimentally measured isotopic yields in some of the foils, it did not fully resolve the observed discrepancy for the entire foil target stack. This paper describes an effort to reconcile measured and computational data.

2. Materials and methods

The ^{27}Al(p,x)^{22}Na reaction has a well-characterized energy-dependent production cross section, see Fig. 1, in the energy range of interest, 30–100 MeV. Due to the extent of its characterization compared with other reactions, this reaction is almost exclusively used for proton intensity monitoring purposes at these energies [4,5]. We used IAEA NDS recommended cross sections for the ^{27}Al(p,x)^{22}Na reaction, sourced from the IAEA charged particle database [11]. Since no uncertainties are presently assigned to IAEA recommended cross sections, we adopted uncertainties based on the data reported by Steyn et al. [10] as a conservative measure.

Sodium-22 has a well-defined gamma peak at 1274.5 keV and intensity 99.94%. This gamma was used to quantify the activity of ^{22}Na production experimentally. Predicted yields of ^{22}Na generated in the Al monitor foils were calculated using the aforementioned codes: MCNP6, TRIM, and A&Z, and utilizing the IAEA recommended cross sections. These predictions were compared with experimental measurements of ^{22}Na yields.

Time-of-flight measurements revealed a primary beam energy of 99.1 ± 0.5 MeV. The 0.5 MeV uncertainty is derived from an observed variation during the duration of the measurement, and is not a reflection of the error in the experimental measurement itself. The time-of-flight measurement was performed by tapping into existing beam position monitoring equipment for a frequency domain or phase measurement of the beam's micropulses. The measured value of 99.1 MeV was used in all simulations presented herein.

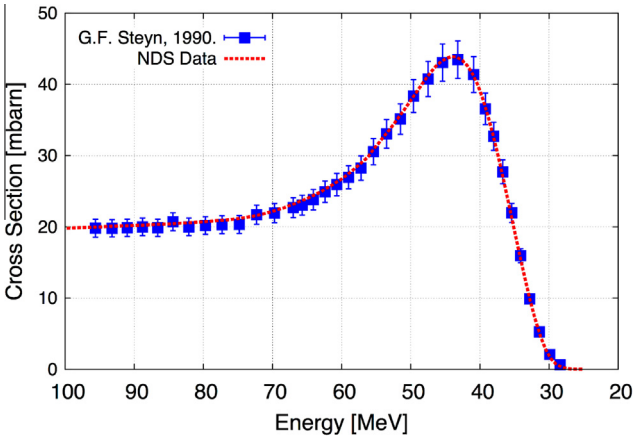

Fig. 1. Published cross section for ^{27}Al(p,x)^{22}Na used in Al monitor foils, from NDS data [11] and from Steyn et al. [10].

2.1. Experimental approach

For the purpose of monitoring the proton fluence in the measurement of Tb + p excitation functions, thin aluminum monitor foils (0.25 mm thickness, 25 x 25 mm) of high purity (Goodfellow Metals, mass 0.417 ± 0.003 g) were encapsulated in 25 μm thick Kapton® tape and stacked with the terbium target foils. To facilitate cross section measurements on terbium at predetermined energies, target foils and their accompanying monitor foils were separated by additional aluminum degraders, included in the design of a specially fabricated aluminum foil holder. The Kapton® enclosed Tb target foils were stacked with the Al foils and irradiated with protons at 100 nA, see Fig. 2.

Following a one hour irradiation, each Al foil was assayed using γ-spectroscopy on calibrated HPGe detectors in the Chemistry Division countroom at LANL. Peaks were identified and corrected for background using the SPECANL analysis algorithm. Details of the experimental activity determination and associated uncertainties have been reported previously [5].

2.2. Computational simulations

The algorithm of the MCNP6 code designed to track the number of protons that cross each aluminum foil is the F4 volumetric tally. In order to obtain an effective fluence at each foil position, the dimensionless MCNP6 tally representing the predicted number of protons was normalized, then multiplied by the fluence calculated from the measured activity for the first foil. Eq. (1) was used to calculate the fluence of the first foil from the measured activity [11].

$$\sigma_i(E) = 2.678 \cdot 10^{-10} \left[\frac{A\lambda N_i}{I\rho x(1-e^{\lambda t})}\right] \quad (1)$$

where $\sigma_i(E)$ is the cross section for the process, [mbarn], A is the atomic mass of the target, [amu], N_i is the is the number of product nuclei present at End-of-Bombardment, I is the average beam current, [μA], ρ is the density of the target material, [g/cm^3], x is the target thickness, [cm], λ is the decay constant, [s^{-1}], t is the irradiation time, [s].

As the beam passes through target material, there is also a statistically driven broadening of its effective energy, the shape of which is predicted by MCNP6 and TRIM. To account for the energy distribution, we calculated an effective or weighted cross section. It is especially important to address energy broadening in regions where the excitation function under consideration varies rapidly with energy. In the excitation function shown in Fig. 1, a strong variation in the energy range from 30 to 65 MeV is observed, the energy region covered by the last 3 foils in the stack.

A correction must be applied to the cross sections to account for energy straggling incident on the foil. An effective cross section for each foil is derived by weighting the excitation function by the distribution of energies incident on the foil. The incident energy takes a Gaussian shaped distribution, $w_i(E_i)$, with the tallies for each energy bin (E_i) determining the parameters of the fit:

$$w_i(E_i) = \frac{1}{\sigma_{SD}\sqrt{2\pi}} \cdot \exp\left[-\frac{(E_i-\mu)^2}{2\sigma_{SD}^2}\right] \quad (2)$$

where μ is the mean energy and σ_{SD} is the standard deviation from the mean energy. The Gaussian distribution was multiplied by corresponding cross sections (σ_i) for each energy bin in order to find a weighted average of the cross section, σ_{wav}: