Measurement and validation of the cross section in the FLUKA code for the production of 63Zn and 65Zn in Cu targets for low-energy proton accelerators postprint

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Abstract

Induced radioactivity is one of the essential problems in the radiation protection field of proton accelerators. Research on the induced radioactivity of low-energy proton accelerators is highly limited. Given such context, this study investigates the cross section of 63Cu (p, n) 63Zn and 65Cu (p, n) 65Zn in Cu targets at 11 MeV proton accelerators through an activation experiment. The uncertainties of the results are analyzed in detail. Results show that the cross section of the reaction of 65Cu (p, n) 65Zn in the experiment is only 1.36% lower than that of the FLUKA simulation, whereas the reaction of 63Cu (p, n) 63Zn in the experiment is 25.4% higher than that of the FLUKA simulation. Given that the benchmark for the FLUKA code of low-energy proton accelerators is very limited, this study provides a reference in this field.

Full Text

Preamble

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Measurement and validation of the cross section in the FLUKA code for the production of ^{63}Zn and ^{65}Zn in Cu targets for low-energy proton accelerators

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Induced radioactivity is one of the essential problems in the radiation protection field of proton accelerators. Research on the induced radioactivity of low-energy proton accelerators is highly limited. Given this context, this study investigates the cross sections of ^{63}Cu(p,n)^{63}Zn and ^{65}Cu(p,n)^{65}Zn reactions in Cu targets at 11 MeV proton accelerators through activation experiments. The uncertainties of the results are analyzed in detail. Results show that the cross section for the ^{65}Cu(p,n)^{65}Zn reaction from the experiment is only 1.36% lower than that from FLUKA simulation, whereas the cross section for the ^{63}Cu(p,n)^{63}Zn reaction from the experiment is 25.4% higher than that from FLUKA simulation. Given that benchmarks for the FLUKA code at low-energy proton accelerators are very limited, this study provides a valuable reference in this field.

Keywords: Activation analysis experiment, Cross section measurement, Induced radioactivity, FLUKA, Copper target
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INTRODUCTION

The radiation protection of proton accelerators is a critical issue for the safety of both personnel and the public. One particular concern at proton accelerators is induced radioactivity, as intense primary proton beams, secondary neutron beams, and scattered particles can activate the materials of accelerator components. From a radiation safety perspective, quantitatively evaluating the radioactivity induced in different materials is essential. Monte Carlo simulation or theoretical analysis methods may be employed to estimate induced radioactivity.

FLUKA is a general-purpose Monte Carlo code that calculates particle transport and their interactions with matter, applicable across a broad range of applications from proton and electron accelerator shielding to target design, calorimetry, activation, dosimetry, detector design, accelerator-driven systems, cosmic rays, neutrino physics, radiotherapy, and more. FLUKA is widely used to estimate the induced radioactivity of accelerator components, as well as in tunnel air and cooling water, for personnel radiation protection. Benchmark experiments have been conducted to verify FLUKA's accuracy in calculating induced radioactivity at high-energy proton and electron accelerators. However, studies comparing FLUKA simulations with experimental data for low-energy proton accelerators remain scarce.

Investigating induced radioactivity at low-energy proton accelerators is important given their wide range of applications. For example, medical proton accelerators that produce radioactive isotopes typically operate between 10 MeV and 18 MeV. In the present study, the cross sections of ^{63}Cu(p,n)^{63}Zn and ^{65}Cu(p,n)^{65}Zn reactions in Cu targets at an 11 MeV proton accelerator were investigated through activation experiments and Monte Carlo simulation. Copper was chosen because it is frequently used in proton accelerator structures and is relatively pure compared to other materials. The isotopes ^{63}Zn and ^{65}Zn, produced by the ^{63}Cu(p,n)^{63}Zn and ^{65}Cu(p,n)^{65}Zn reactions respectively, were investigated because they are the two most important isotopes generated in Cu targets. The measured results and corresponding FLUKA simulations were compared to verify FLUKA's accuracy in the low-energy range for proton accelerators.

II. METHODS

The activation analysis method, which originated in the 1930s, is widely used in many fields. The method is described in the subsequent sections.

Suppose a thin target sample containing element N_1 is placed in the homogeneous flux of a proton beam. The incident protons induce the nuclear reaction N_1(p,n)N_2. The cross section σ is expressed as:

$$\sigma = \frac{\lambda N_{\text{irr}}}{I n_d (1 - e^{-\lambda t_{\text{irr}}})}$$

where λ is the decay constant of the activated nuclei; I is the current of the incident beam (i.e., incident particle counts per unit time, s^{-1}); n_d is the number of target nuclei per unit area with thickness d (cm^{-2}), given by:

$$n_d = \frac{\rho d N_A}{m}$$

where is the target density in g/cm^3; m is the atomic mass of the target material; and N_A is Avogadro's number.

In the experiment, N_{irr} is derived using the following formula:

$$N_{\text{irr}} = \frac{N_{\text{decay}}}{e^{-\lambda t_1}(1 - e^{-\lambda t_{\text{decay}}})}$$

where t_1 is the cooling time after irradiation ends, t_{decay} is the duration of the activity measurement, and N_{decay} is the number of decays measured in the radioactive nuclei.

III. EXPERIMENT

A. Target Properties

The Cu targets used in the experiment had a disc shape with a diameter of 1.7 cm and thickness of 30 µm, according to national standard precision requirements. The major properties of the targets are listed in Table 1 . Three Cu targets were used: one for a pre-test to establish the experimental procedures, and two for cross section measurements, labeled as samples 1 and 2.

Table 1. Properties of the target used in experiment

Isotope	Atomic percent(%)	Daughter	Half-life
^{63}Cu	69.17	^{63}Zn	38.47 min
^{65}Cu	30.83	^{65}Zn	244.2 days

B. Irradiation of the targets

The targets were irradiated using an RDS 111-type proton cyclotron. The proton energy was 11 MeV, and the beam spot diameter was less than 2 mm. The range of 11 MeV protons in copper is approximately 386.5 µm, which is much larger than the target thickness. The beam intensity was easily adjusted and measured using a Faraday cup, and was set to several µA during the experiment. The irradiation time for each sample was 4 s and was controlled manually through the accelerator's control software.

Figure 1 [Figure 1: see original paper] shows that the beam was defined by a collimator. The incident beam intensity was measured with a Faraday cup mounted directly behind the target. The stability of the beam intensity was monitored with a current integrator. The corresponding irradiation intensities for samples 1 and 2 were 2.66 µA and 3.02 µA, respectively.

C. Measurement of the induced radioactivity

After irradiation, the activated samples were placed in a separate low-background counting area and measured using a hyperpure germanium detector. The energy resolution (FWHM) of this GEM70-S gamma spectrum detector was 1.90 keV at 1332.5 keV. The energy spectrum data were obtained through 8192 channels of a digital spectrometer (DSPEC Plus). The entire system was calibrated using the virtual calibration software Gamma Vision 32 just before measurement. The detection efficiencies for different photon energies are shown in Fig. 2 [Figure 2: see original paper].

Fig. 2. Detection efficiencies for different photon energy.

Given the statistical error requirements for the energy spectrum measurement, approximately 1.5 h was required to measure the decay spectrum of ^{63}Zn, and approximately 24 h for that of ^{65}Zn. Another reason for this difference is that the half-life of ^{63}Zn (38.5 minutes) is much shorter than that of ^{65}Zn (243.7 days). The activities of ^{63}Zn and ^{65}Zn at the stoppage time were obtained from the product of the detection efficiency and N_{irr}, where N_{irr} is calculated using Eq. (3).

Table 2 lists the counting results, detection efficiency, and activities of ^{63}Zn and ^{65}Zn at the stoppage time, along with their corresponding errors for samples 1 and 2. The errors mainly originated from statistical counting errors.

IV. MONTE CARLO SIMULATION WITH FLUKA

The cross section data in FLUKA were partially obtained from the experimental database of the American National Nuclear Data Center. The proton and neutron inelastic cross sections between 10 and 200 MeV were updated by fitting them to experimental data. An accurate treatment of the cross-section energy dependence for all charged particles, independent of step size, was introduced at that stage through the fictitious-σ method. The present treatment for hadron-nucleus cross sections was based on a novel approach that combined experimental data, data-driven theoretical approaches, PDG fits, and phase-shift analysis when available.

To compare the measured cross section results with the corresponding data used in the FLUKA code, a detailed Monte Carlo simulation of the experiment was also conducted. The parameters in the simulation were identical to those in the experiment, including beam energy, geometry, and irradiation profile. The simulation was based on a detailed description of the experimental setup, with samples defined using their actual dimensions.

The full hadronic cascade was simulated in the experimental setup. Neutrons were transported down to thermal energies, and a threshold of 1 keV was applied to all other hadrons. Several physical settings were activated, including the PEANUT model at all energies, treatment of coalescence, and evaporation of heavy fragments. The activities of the radionuclides ^{65}Zn and ^{63}Zn in the samples at a certain cooling time were calculated using FLUKA. The cross sections of ^{65}Cu(p,n)^{65}Zn and ^{63}Cu(p,n)^{63}Zn used by FLUKA were then deduced using Eq. (1). The error in the cross section was mainly caused by statistical error in the activity calculation, which was 0.1% for ^{65}Zn and 0.2% for ^{63}Zn. The relative uncertainties of the cross sections from the simulation were 0.1% and 0.2% for ^{65}Cu(p,n)^{65}Zn and ^{63}Cu(p,n)^{63}Zn, respectively.

V. RESULTS AND DISCUSSION

A. Uncertainty analysis of the cross section

To analyze the discrepancy between the measured cross section and that from the FLUKA simulation, a detailed uncertainty analysis was conducted on the measurement results. The uncertainty of the cross section data from the FLUKA simulation was mainly due to statistical error in the calculation.

Equation (1) indicates that the uncertainty in the results originated from errors in the target nuclei number density (n_d), activity at stoppage time (A_{irr}), beam intensity (I), and irradiation time (t_{irr}). The formula is given by:

$$\sigma_\sigma = \sigma \left[\left(\frac{\sigma_{A_{\mathrm{irr}}}}{A_{\mathrm{irr}}} \right)^2 + \left(\frac{\sigma_{n_d}}{n_d} \right)^2 + \left(\frac{\sigma_I}{I} \right)^2 + \left(\frac{\lambda e^{-\lambda t_{\mathrm{irr}}}}{1 - e^{-\lambda t_{\mathrm{irr}}}} \sigma_{t_{\mathrm{irr}}} \right)^2 \right]^{1/2}$$

where σ is the cross section, and $\sigma\sigma$, σ{Airr}, *σ{nd},* σI, and *σ{irr} are the standard deviations of σ, A*{irr}, n_d, I, and t_{irr}, respectively.

The uncertainties can be classified as follows:

a) Errors in target manufacture
These include errors in thickness, purity, and inhomogeneity of the thin copper foil. According to references [21, 22], the difference in the quantities of ^{63}Cu and ^{65}Cu in natural Cu at different locations is small, generally less than 0.2%. Therefore, the influence of isotope abundance can be neglected. Given that machining techniques for creating 30 µm thick Cu targets are very mature, errors attributed to purity and inhomogeneity of the thin copper foil were negligible compared to errors in target thickness. Consequently, the uncertainty in target manufacture originated mainly from thickness uncertainty. According to the manufacturer, the standard deviation of the thickness was 0.003 mm. With a Cu foil thickness of 0.03 mm, the relative uncertainty is approximately 10%.

b) Uncertainties in the irradiation period
These include errors in proton beam energy, irradiation time, and beam intensity. The beam energy of the RDS 111-type proton cyclotron was 11 MeV, and the energy uncertainty was found to be negligible based on measurements provided by the accelerator control department. As previously mentioned, the irradiation time for each sample was 4 s, with an estimated error within 0.5 s. The error in beam intensity was estimated to be 0.01 µA.

c) Uncertainties in the measurement of induced radioactivity
These include errors in detection efficiency, counting time, and statistical counting errors. The virtual calibration software Gamma Vision 32 specification for the GEM70-S gamma spectrum detector shows that the uncertainty in detection efficiency was lower than 2.5%. The uncertainty in counting time was very small and negligible. The counting results were typically higher than 10^4 (i.e.,

the counting errors are less than 3% with a confidence interval of 99.7%). According to Eq. (2), the relative uncertainty of N_{irr} at the 99.7% confidence interval is given by:

$$R_{N_{\mathrm{irr}}} = \left[R^2_{N_{\mathrm{decay}}} + (2.5\%)^2\right]^{1/2}$$

where the uncertainties of the decay constant and counting time are neglected.

The activity at the stoppage time of irradiation, A_{irr}, can be derived using:

$$A_{\mathrm{irr}} = \lambda N_{\mathrm{irr}}$$

The relative uncertainty of A_{irr} can then be calculated by:

$$R_{A_{\mathrm{irr}}} = R_{N_{\mathrm{irr}}} = \left[R^2_{N_{\mathrm{decay}}} + (2.5\%)^2\right]^{1/2}$$

The results for the relative uncertainty of A_{irr} are shown in Table 2. The uncertainties of the measured cross sections were obtained from the preceding discussion and are listed in Table 3.

B. Comparison of cross sections

Table 3 summarizes the comparison of experimental results, FLUKA simulation results, and other published data based on different calculation models [22–25] for the cross sections of ^{63}Cu(p,n)^{63}Zn and ^{65}Cu(p,n)^{65}Zn reactions. The uncertainties of the results were discussed in detail in the previous sections.

Both the average cross sections and FLUKA simulation results are within the range of various results from several references, which were obtained from theoretical calculations. For the ^{65}Cu(p,n)^{65}Zn reaction, the difference between the experimental and FLUKA simulation cross sections was only 1.36%. However, for the ^{63}Cu(p,n)^{63}Zn reaction, the cross section measured in this work was 25.41% higher than that in the FLUKA simulation.

Table 3. Reaction cross sections of ^{63}Cu(p,n)^{63}Zn and ^{65}Cu(p,n)^{65}Zn (mb)

Reaction	Sample 1	Sample 2	Average	FLUKA
^{63}Cu(p,n)^{63}Zn	391\$±63|375±60|383±4|302±\$5			
^{65}Cu(p,n)^{65}Zn	674\$±108|640±102|657±72|658±\$15			

VI. CONCLUSIONS

Induced radioactivity is one of the essential problems in the radiation protection of proton accelerators. The accuracy of estimating induced radioactivity relies on the precision of cross section data. Therefore, verifying related cross section data through simulation and experiment is important, particularly since data for low-energy protons are limited. In this work, the cross sections of the ^{65}Cu(p,n)^{65}Zn and ^{63}Cu(p,n)^{63}Zn reactions were studied using both the activation analysis method with irradiation experiments and detailed Monte Carlo simulation with the FLUKA code. The uncertainties of the results were also analyzed in detail.

The cross sections obtained in this work through both experiment and simulation are within the range of results from published papers, which were mainly based on theoretical calculations. A slight difference of 1.36% was observed between the experimental and FLUKA simulation cross sections for the ^{65}Cu(p,n)^{65}Zn reaction. However, the experimental data for the ^{63}Cu(p,n)^{63}Zn reaction was 25.4% higher than the FLUKA simulation results. Given that benchmarks for the FLUKA code at low-energy proton accelerators are limited, this work provides a valuable reference in this field.

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Note: Figure translations are in progress. See original paper for figures.