Measurement of neutron yields from a thick uranium target bombarded by high-energy 4He ions

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Abstract

The neutron yields induced by 500 MeV/u 4He ions bombarding a thick uranium target have been measured by the time-of-flight method at Heavy Ion Research Facility in Lanzhou. Simulations employing three Monte Carlo codes, GEANT4, FLUKA, and PHITS, were performed to calculate neutron yields under conditions identical to the experiment. The results indicate that all three codes reproduce key features of the experimental energy spectrum distribution to varying degrees, though each exhibits distinct advantages and limitations across specific energy intervals and spatial angles. This study provides critical data for optimizing the spallation target and subcritical reactor designs in accelerator-driven subcritical systems.

Full Text

Preamble

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The neutron yields induced by 500 MeV/u 4He ions bombarding a thick uranium target have been measured by the time-of-flight method at the Heavy Ion Research Facility in Lanzhou. Simulations employing three Monte Carlo codes—GEANT4, FLUKA, and PHITS—were performed to calculate neutron yields under conditions identical to the experiment. The results indicate that all three codes reproduce key features of the experimental energy spectrum distribution to varying degrees, though each exhibits distinct advantages and limitations across specific energy intervals and spatial angles. This study provides critical data for optimizing spallation target and subcritical reactor designs in accelerator-driven subcritical systems.

Keywords: Thick target neutron yield, Helium ion, Uranium, GEANT4, FLUKA, PHITS

Introduction

Nuclear energy has emerged as a critical component of global energy transition, attributed to its high energy density, zero-carbon emission profile, and stable power supply [1–5]. However, as the installed capacity of nuclear power plants expands, the accumulated spent fuel in China is projected to exceed 20,000 tons by 2030 [6]. If not properly managed, the spent fuel may pose a significant threat to the ecological environment and human health through groundwater infiltration. The safe management of spent fuel has become a key constraint on the long-term sustainability of nuclear fission energy systems. Specifically, the minor actinides (MAs) and long-lived fission products (LLFPs) present in spent fuel pose a long-term radiological hazard to humanity, as their radioactivity requires tens of thousands to millions of years to decay to the level of natural uranium ore [7, 8].

To fundamentally address the radiological hazards of spent fuel, it is essential to transmute the MAs and LLFPs. This need has consequently driven the development of the Partitioning & Transmutation (P&T) strategy [9, 10]. Since the early 1990s, with advancements in high-current accelerators, a novel waste treatment approach—accelerator-driven subcritical system (ADS) based P&T—has been proposed and extensively investigated [11–13]. The ADS comprises three core components: a high-current accelerator, a spallation target, and a subcritical reactor. In this system, high-energy neutrons generated by bombarding a heavy-metal spallation target with a primary particle beam transmute the MAs and LLFPs in the subcritical reactor [14–17]. Relevant studies indicate that the required storage time for the transmuted spent fuel can be reduced to several centuries [18]. Additionally, high-energy neutrons can induce 238U to participate in chain reactions, significantly enhancing the utilization of uranium resources. Collectively, ADS integrates spent fuel transmutation with efficient energy production, offering a novel pathway for the sustainable development of nuclear energy [19, 20].

The transmutation of long-lived waste in ADS depends on high-energy spallation neutrons. Accurate acquisition of nuclear data, particularly the neutron yields and angular distribution resulting from high-energy beam interactions with the spallation target, is essential for optimizing ADS neutron source design, including target material selection and accelerator energy matching [21–24]. However, the absence of high-energy nuclear data related to spallation reactions and the uncertainty of existing datasets pose major constraints on ADS engineering. Studies indicate that current data uncertainties can propagate to effective multiplication factor calculations for subcritical reactors with errors up to 2.77%, thereby compromising predictions of the core power profile and assessments of the safety margin [25]. Consequently, high-precision nuclear data measurement is an essential prerequisite for enhancing both the economic viability and operational safety of ADS designs.

Current studies about ADS spallation targets primarily focus on proton-driven schemes. Systematic investigations of multi-nucleon projectiles such as 4He ions remain limited. 4He ions have a higher mass number compared to protons, which might generate a denser neutron flux during spallation reactions. Meanwhile, 4He-induced reactions yield harder neutron energy spectra than those from protons, which is favorable for transmuting long-lived waste [26]. Despite these advantages, 4He application in ADS is constrained by critical nuclear data deficiencies, notably the neutron yields from high-energy 4He interactions. In addition, uranium serves as both a component of fuel assemblies and a potential spallation target material for ADS. Uranium-related high-energy nuclear reaction parameters, such as neutron yields from 4He + 238U interactions, provide critical guidance for optimizing neutron source design [27–33].

This study experimentally measured the neutron yields from a thick uranium target bombarded by 500 MeV/u 4He ions, with measurements complemented by GEANT4 [34, 35], FLUKA [36] and PHITS [37] Monte Carlo simulations. The dual approach aimed to identify key characteristics of neutron energy and spatial distributions. The results will provide essential input parameters for ADS neutron source design, enable efficient transmutation of MAs and LLFPs, and expand the available primary beam options.

II. Experiment

A. Experimental Setup

The Heavy Ion Research Facility in Lanzhou (HIRFL) comprises an electron cyclotron resonance (ECR) ion source, a sector focus cyclotron (SFC), a separated sector cyclotron (SSC) and a cooler-storage-ring (CSR) system [38, 39]. The CSR system constitutes a dual-cooler storage ring complex, consisting of a main ring (CSRm), an experimental ring (CSRe), and a radioactive beam line interconnecting the two rings [40]. Ions from SFC and SSC, in the energy range of 2–25 MeV/u, are injected, accumulated, and accelerated in CSRm to achieve higher energies of 100–500 MeV/u. These accelerated beams are subsequently extracted for radioactive ion beam production or generation of highly charged heavy ions. The secondary ion beams can be captured and stored in CSRe for internal target experiments or high-precision spectroscopic studies utilizing electron cooling technology. Alternatively, beams can be delivered through slow extraction from CSRm for external target experiments or applications in cancer therapy.

The 500 MeV/u 4He beam used in this experiment was delivered by the Proton Induced Spallation (PISA) terminal of the HIRFL-CSR facility. A schematic of the experimental setup is presented in Fig. 1 [Figure 1: see original paper]. Two thin plastic scintillator detectors (BM1 and BM2) were employed for real-time beam monitoring. The signals from BM detectors triggered the data acquisition system and were used to count incident particles per run.

The uranium target, constructed from stacked uranium rods, was a cuboid whose thickness, width and height were 14 cm, 30 cm and 10.5 cm respectively. The target had sufficient thickness to completely stop the primary 4He beam. The range of the 4He beam in the uranium target was calculated using LISE++ [41], revealing the range to target thickness ratio of approximately 0.895.

The liquid scintillator detector array was positioned at 0°, 12°, 30°, 45°, and 60° relative to the beam direction in the laboratory frame. Each liquid scintillator was covered by a thin plastic scintillator which functioned as a veto detector to discriminate charged particles generated in the target.

To quantify laboratory background neutron contributions, measurements were performed for each detector configuration using shadow bars. These bars fully blocked particles traveling directly from the target to the liquid scintillators, ensuring that any neutron events recorded in the detectors were attributed to background.

B. Electronic Equipment and Data Acquisition

The simplified electronic device architecture used for the experiment is presented in Fig. 2 [Figure 2: see original paper]. The start scintillator signals BM1 and BM2 are passed through a splitter to form a two-channel processing path: one is fed into a charge-to-digital converter (QDC), and the other is connected to a constant fraction discriminator (CFD) for signal conditioning. After being shaped by the CFD, the two signals are combined by a logic AND gate and subsequently assigned to three independent processing channels. The first channel is directly connected to the scaler module for counting the number of particles incident per single pulse; the second channel is connected to the time-to-digital converter (TDC) after conditioning by the time delay unit to serve as the termination trigger signal for time-of-flight (TOF) measurements; and the third channel is connected to coincide with the output signal of the liquid scintillator to generate the main trigger signal.

The output signal of the liquid scintillator detector is processed in a three-branch scheme: the first branch is connected to the TDC module as the start trigger signal for the TOF measurement; the remaining two branches are connected to the QDC system with different gate width parameters. Specifically, the total gate width channel is used to collect the full integral of the signal of the scintillator light output, while the slow gate width channel mainly captures the tail component of the light decay curve. In the subsequent data processing, pulse shape discrimination (PSD) is realized by calculating the charge difference between the two gate width channels, thereby enabling the differential identification of neutron and photon events.

In addition, the veto detector output signal is connected to the QDC module to reject interference from charged particle events.

III. Data Filtering

The experimental data were recorded by the data acquisition system and subsequently analyzed using the ROOT framework [42]. Among the secondary particles produced by 4He ions, only neutrons were of interest in this study. To extract valid neutron events information, a series of filters were designed, including charged particle events removal and neutron-photon events discrimination. Following effective neutron identification, the thick target neutron yields were determined via the TOF method.

A. Charged Particle Discrimination

Charged particle discrimination was implemented by exploiting the distinct interaction characteristics of charged particles and neutral particles in plastic scintillators. Neutral particles deposit much less energy in the veto detector than charged particles, with their signals confined within the low-channel peak, whereas charged particle events are located on the right side of the peak. These features are illustrated in the QDC spectrum of the 0° veto detector (Fig. 3 [Figure 3: see original paper]). A Gaussian distribution was fitted to the peak, and a 3σ cutoff threshold was set at the right of the fitted center. This configuration effectively discriminates charged particle events while preserving potential neutron candidates for subsequent yield calculations.

B. Neutron-Photon Discrimination

After separating neutral and charged particles, further discrimination between neutrons and photons is necessary. Organic liquid scintillators possess outstanding PSD capability. The underlying physical mechanism arises from distinct scintillation light components: neutron interactions predominantly generate recoil protons via nuclear collisions, producing signals with higher slow-component fractions, while photon events create Compton electrons through atomic electron interactions, characterized by signals dominated by fast components. This fundamental difference is manifested in pulse waveform characteristics, as quantified by the two-dimensional PSD spectrum comparing total component integration (Qtotal) versus slow component integration (Qslow), and shown in

Fig. 4 [Figure 4: see original paper]. A clear separation between neutron and photon events was achieved through optimized charge integration gate settings, enabling precise identification of neutron signals.

C. Thick Target Neutron Yields Calculation

With the purified neutron TDC spectrum obtained, neutron energy was determined by TOF method. The start signal was generated by each liquid scintillator detector, and the stop signal was provided by the coincidence output of BM1 and BM2 signals. The neutron energy En was calculated using Eq. (1):

$$E_n = \frac{m_n c^2}{2}\left(\frac{L}{c\Delta t_{n-\gamma}}\right)^2$$

where m_n denotes the neutron rest mass, c is the speed of light in vacuum, $\Delta t_{n-\gamma}$ represents the TOF difference between prompt γ-rays and neutrons, and L is the neutron flight distance. The flight distances from the target center to detectors at 0°, 12°, 30°, 45°, and 60° were 480 cm, 400 cm, 300 cm, 300 cm, and 250 cm, respectively, with positional uncertainties below 0.5% after laser alignment.

The energy resolution can be estimated by Eq. (2):

$$\frac{\Delta E}{E} = \frac{\gamma(\gamma+1)}{(\gamma-1)}\frac{\Delta t}{t}$$

where $\Delta E/E$ denotes the energy resolution, γ is the Lorentz factor, t is the neutron TOF, and Δt corresponds to the time resolution. The time resolution is determined by the full width at half maximum (FWHM) of the prompt γ-rays peak in the TDC spectrum. The time resolution incorporates contributions from the intrinsic detector time resolution, the inherent beam time structure, and variations in the interaction position within the liquid scintillator detectors.

The thick target double differential neutron yields were calculated using Eq. (3):

$$\frac{d^{2Y}}{dEd\Omega} = \frac{N_n}{N_\alpha \epsilon \Delta E \Delta\Omega}$$

where N_n denotes the detected neutron count, N_α is the number of incident 4He ions, ΔE is the energy bin width, $\Delta\Omega$ is the detector solid angle, and ϵ is the neutron detection efficiency of the liquid scintillator detectors. In this study, the SCINFUL-QMD code was employed to estimate the neutron detection efficiency [43], and it has been benchmarked in previous studies. For neutron energies below 150 MeV, SCINFUL-QMD employs an extended library interface to calculate detection efficiency. For neutron energies above 150 MeV, it combines quantum molecular dynamics with a statistical decay model and incorporates 39 potential neutron reaction channels to derive the results. The benchmarked SCINFUL-QMD code demonstrates reliable prediction capability for neutron detection efficiency across an extended energy range from 10 keV to 3 GeV, as verified through systematic validation studies. The neutron detection efficiencies used in this study are given in Fig. 5 [Figure 5: see original paper].

D. Uncertainty Analysis

The uncertainties in the measurement data are primarily attributed to uncertainties in the simulated detection efficiency, statistical errors in detector counts, and deviations in the solid angle acceptance. Previous studies indicate that the detection efficiency uncertainty calculated by SCINFUL-QMD is approximately 10%.

In the medium-to-low energy regions of the neutron yield spectra, the statistical error in counts is maintained below 10%, but increases to 15–25% in the high-energy region. The uncertainty in solid angle acceptance arises from variations in the interaction positions of primary particles with the target and in the reaction positions of secondary particles within the scintillator. Even under extreme positional assumptions, this uncertainty remains constrained below 8%.

IV. Results and Discussion

A. Double Differential Neutron Yields

Fig. 6 [Figure 6: see original paper] shows the double differential neutron yield spectrum from bombardment of a thick uranium target by 500 MeV/u 4He ions, comparing experimental measurements with Monte Carlo simulations. The simulations employed GEANT4 11.1.2 and PHITS 3.32, both utilizing the Intra-Nuclear Cascade Liege (INCL) model [44], and FLUKA 4-3.3 with the Relativistic Quantum Molecular Dynamics (RQMD) model [45] to test their accuracy and validity. Curves corresponding to different detection angles are vertically offset for visual clarity.

Low-energy neutrons below 20 MeV primarily originate from evaporation of residual nuclei. The calculations of neutron yields in this energy region rely mainly on established nuclear reaction data, leading to similar simulation results from the three Monte Carlo codes. Among all measured angles, GEANT4 predicts the highest neutron yield, while PHITS gives the lowest. However, all simulations underestimate the measured neutron yields, likely due to the neglect of high-energy neutron induced fission in the uranium target. Quantitatively, at 0°, the average relative errors of three Monte Carlo simulations range from 45% to 65% compared to experimental data. As the angle increases, these errors gradually decrease and stabilize between 13% and 30%.

Intermediate-energy neutrons in the 20–100 MeV range are mainly attributed to the pre-equilibrium emission of compound nuclei. Monte Carlo simulations achieve optimal agreement with experimental measurements in this energy range, accurately reproducing both the magnitude and the trend of the yields. FLUKA provides the closest match to measured neutron yield at 0°, while GEANT4 performs better at other angles. In this energy range, FLUKA demonstrates relatively smaller relative errors in the forward-angle region, with an approximate value of 19%, whereas its errors in the large-angle direction show a progressive increase to around 35%. GEANT4 and PHITS both present errors of approximately 44% at 0°, and their errors rapidly diminish with increasing angle, reaching a minimum of roughly 4% at 60°.

Secondary neutrons with energies above 100 MeV are primarily produced by projectile fragmentation. Although high-energy neutrons are generally sparse, a broad high-energy peak forms at approximately 75% of the projectile energy at 0°. GEANT4 calculation is closest to this high-energy broad peak, with a similar slope and magnitude. Notably, GEANT4 and PHITS yield curves show strong similarity at angles greater than 30°, attributed to their simultaneous use of the INCL model (different versions). This also indicates that updates to the INCL model have primarily optimized the high-energy region in the forward direction, enhancing its predictive capability for high-energy neutrons. At 0°, the PHITS calculation has a large error, while the GEANT4 calculation error remains below 30%. This discrepancy highlights the effectiveness of the INCL model optimization for the direct reaction process. FLUKA also achieves commendable results, with a mean relative error of approximately 48% across all tested configurations.

Multi-angle coverage and multi-code benchmarking provide critical validation data for next-generation radiation transport simulations.

B. Angular Neutron Yields Distribution

The angular distribution of secondary neutrons is analyzed by integrating the neutron double differential yields across the energy dimension at each angle to determine the angular yield. To mitigate contributions from isotropic evaporation neutrons, a 7 MeV energy threshold was applied during angular yield calculations. Fig. 7 [Figure 7: see original paper] (a) compares experimental (EXP) and simulated (GEANT4, FLUKA, PHITS) angular yield distributions, while Fig. 7 [Figure 7: see original paper] (b) presents the C/E (Calculation/Experiment) ratio to quantify simulation accuracy.

Experimental results reveal a distinct angular correlation in secondary neutron emission. The angular yields peak at 0° and decrease gradually from 1.7 n/sr/ion to 0.3 n/sr/ion as the emission angle increases. This distribution confirms the expected forward-peaked emission profile of spallation neutrons. Monte Carlo simulations demonstrate varying degrees of agreement with experimental measurements.

GEANT4 calculations show general consistency with experimental trends, albeit with some deviations. Specifically, GEANT4 significantly underestimates the angular yield at 0°. Meanwhile, the C/E ratios at other angles fluctuate between 0.77 and 1.11. The mean deviation in GEANT4's angular yield is approximately 35.9%.

In contrast, FLUKA simulations exhibit improved agreement with experimental data, particularly at large angles. FLUKA C/E ratios cluster between 0.88 and 1.41, with an average angular yield deviation of 16.5%, indicating superior overall consistency compared to GEANT4. PHITS simulations, however, demonstrate significant overestimation, especially at forward angles, where C/E ratios reach 1.37–1.87. This deviation is due to exaggerated amplitudes of high-energy peaks in forward-angle neutron yield spectra. Notably, PHITS achieves close agreement with experimental values at large angles, with deviations below 5%.

Fig. 8 [Figure 8: see original paper] illustrates the trend of cumulative neutron yields as the polar angle increases from 0° to a specific angle in spherical coordinate space. As shown, the cumulative yields continuously increase with polar angle. The experimental cumulative yield for 60° indicates that approximately 1.57 secondary neutrons (En > 7 MeV) are produced per incident 4He ion within the corresponding solid angle cone.

When compared to experimental data, Monte Carlo simulation results from different codes demonstrate discrepancies. The FLUKA prediction lies above the experimental data overall, with its maximum approximately 7% higher than the measured data. This discrepancy may arise from its conservative treatment of physical processes, leading to overpredicted results. At forward angles, GEANT4 and PHITS calculations are in good agreement with experiments, whereas at large angles, the discrepancies among the three codes gradually increase, reaching a maximum of 11.6%.

These deviations reflect the differences in how the various simulation models handle neutron production, transport and detection response. They can be attributed to the differences in the modeling of physical processes (e.g., nuclear reaction cross section, particle transport mechanism), geometric configurations, and boundary condition settings of the different methods. The results provide valuable insights into neutron spatial distribution and help verify the accuracy of the simulation models. This will facilitate further investigations into the spatial distribution of spallation neutrons and promote advancements in related nuclear physics and radiation transport research.

V. Conclusion

In this study, the neutron yields induced by bombarding a thick uranium target with a 500 MeV/u 4He ion beam were measured using the TOF method at the PISA terminal of HIRFL. The corresponding secondary neutron energy spectrum and angular distribution were obtained. Additionally, secondary neutron yields under identical experimental conditions were simulated using three Monte Carlo codes: GEANT4, FLUKA, and PHITS. The results demonstrate that the three Monte Carlo codes exhibit varying capabilities in reproducing the experimental data. All codes systematically underestimate low-energy neutron contributions across all angular directions, while predictions of high-energy neutron peaks generated by projectile fragmentation in the forward-angle region show inter-code discrepancies. Regarding angular yield distributions, all three codes successfully replicate the strong forward-peaking trend of spallation neutrons, though quantitative deviations from experimental measurements persist. Nevertheless, cumulative neutron yields predicted by all codes remain in an acceptable range.

In summary, investigating neutron yields from high-energy 4He ion bombardment on a thick uranium target is essential for advancing ADS technology. In this study, reliable neutron yield data were obtained by combining experimental measurements with theoretical simulations. These data support the design and construction of the ADS and contribute to the sustainable development of nuclear fission energy.

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Author Contributions

All authors participated in the conception and execution of the experiments. Material preparation, data collection and analysis were performed by Ze-Kun Zhang and Rui Han. Theoretical calculation and analysis were performed by Ze-Kun Zhang and Zhi-Qiang Chen. The first draft of the manuscript was written by Ze-Kun Zhang, and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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