Measurement and calculation of leakage neutron spectra from 238U induced by broad-spectrum neutrons

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Abstract

Leakage neutron spectra from a pure 238U slab sample (dimensions: 100 mm × 100 mm × 20 mm) bombarded by broad-spectrum neutrons were measured at 0° using the time-of-flight method. The experiment was carried out at the Radioactive Ion Beam Line of the Heavy Ion Research Facility in Lanzhou at the Institute of Modern Physics, Chinese Academy of Sciences. Broad-spectrum neutrons were generated by bombarding a tungsten target with 80.5 MeV/u 12C ions. Additionally, calculations were performed in GEANT4 with the INCL, BIC and BERT physics models, in combination with the evaluated nuclear data libraries ENDF/BVIII, JEFF-3.3, and JENDL-4.0, and the theoretical results for the leakage neutron spectrum of the 238U were obtained under the same experimental conditions. The results indicate that the GEANT4 calculations can reasonably reproduce the experimental data.

Full Text

Measurement and Calculation of Leakage Neutron Spectra from ²³ U Induced by Broad-Spectrum Neutrons

H. Sun¹², † X. Zhang¹², Z. Chen¹², R. Han¹², ‡ B. Yang¹, S. Khasanov³, P. Zhang¹ , G. Tian¹², B. Liu¹², F. Shi¹², Z. Zhang¹ , Q. Li¹ , and P. Luo¹²

¹Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China
²Gansu Isotope Laboratory, Lanzhou, 730300, China
³Samarkand State University, Samarkand 140104, Uzbekistan
 School of Nuclear Science and Technology, University of Chinese Academy of Sciences, Beijing 100049, China

Leakage neutron spectra from a pure ²³ U slab sample (dimensions: 100 mm × 100 mm × 20 mm) bombarded by broad-spectrum neutrons were measured at 0° using the time-of-flight method. The experiment was carried out at the Radioactive Ion Beam Line of the Heavy Ion Research Facility in Lanzhou at the Institute of Modern Physics, Chinese Academy of Sciences. Broad-spectrum neutrons were generated by bombarding a tungsten target with 80.5 MeV/u ¹²C ions. Additionally, calculations were performed in GEANT4 with the INCL, BIC, and BERT physics models, in combination with the evaluated nuclear data libraries ENDF/B-VIII, JEFF-3.3, and JENDL-4.0, and the theoretical results for the leakage neutron spectrum of the ²³ U were obtained under the same experimental conditions. The results indicate that the GEANT4 calculations can reasonably reproduce the experimental data.

Keywords: Leakage neutron spectra, ²³ U, Broad-spectrum neutrons, GEANT4

INTRODUCTION

The Accelerator-Driven System (ADS) is a coupled system comprising a subcritical reactor and an external proton accelerator [?]. The system possesses strong transmutation capabilities, significant neutron economy, and reliable safety performance. Not only can it convert spent nuclear fuel into short-lived isotopes, but it also has the potential to both proliferate nuclear fuel and generate electricity in an efficient and sustainable manner [?, ?]. Therefore, the ADS exhibits favorable energy economics and is an important research area within the field of nuclear energy [?, ?]. The advancement of ADS is currently at a critical stage of technological development [?], necessitating substantial support from nuclear data resources [?]. ²³ U is a critical nuclear material for ADS, and the neutron transport nuclear data of ²³ U form an important foundation for both the ADS and advanced nuclear fuel cycle systems [?]. However, significant discrepancies exist among the evaluated neutron data for ²³ U within the energy range below 20 MeV, and such data are notably scarce in the medium- and high-energy

ranges [**?**]. At present, the existing neutron evaluation data for ^{23} U are difficult to directly apply under the complex conditions of high neutron flux and broad energy spectrum encountered in ADS. Therefore, there is a critical need for detailed and reliable experimental measurements as well as theoretical calculations concerning the relevant nuclear data for the interaction between medium- and high-energy neutrons with ^{23} U.

^{23} U is an extremely critical nuclear material for the Accelerator-Driven System. The content of ^{23} U in spent nuclear fuel is more than 95 percent, and the proportion of reactions involving spallation neutrons and ^{23} U within the reactor core is high, and these reactions are notably complex. The neutron transport processes in ^{23} U serve as an essential basis for ADS [**?**, **?**]. After capturing a neutron, ^{23} U can be transformed into a fissionable nuclide, ^{23} Pu (^{23} U + n $\rightarrow$ ^{23} U $\rightarrow$ ^{23} Np $\rightarrow$ ^{23} Pu fissile) [**?**, **?**]. The recycling of ^{23} U through the uranium-plutonium cycle plays a significant role in enhancing the disposal capacity for spent nuclear fuel and promoting the efficiency of the nuclear fuel cycle. This is also an important means to achieve the sustainable development of nuclear fission energy. In addition, the complex reactions between broad-spectrum neutrons and ^{23} U will affect the neutron characteristics in the spallation target region. In conclusion, accurate and reliable experimental measurements of nuclear data related to broad-spectrum neutron transport in ^{23} U targets ensure the safety of ADS design, construction, and subsequent operation and maintenance. These measurements constitute an indispensable component of ADS development. Additionally, experimental data are vital for verifying the reliability of neutron transport calculation software. Therefore, reliable experimental and calculation studies on the neutron transport process of ^{23} U are required.

In this work, we utilized ^{12}C + W as the neutron source. The leakage neutron spectrum is measured by time-of-flight method, by bombarding the ^{23} U sample target with broad-spectrum neutrons. Moreover, this study employs GEANT4 [**?**] to simulate the experimental process in combination with the INCL++ [**?**], BIC [**?**], and BERT [**?**, **?**] physical models and the ENDF/B-VIII [**?**], JEFF-3.3 [**?**], and JENDL-4.0 [**?**] evaluation databases.

METHODS

A. Experimental Methodology

The experiment was carried out at the Radioactive Ion Beam Line in Lanzhou (RIBLL) [**?**, **?**]. The primary beam was 80.5 MeV/u ^{12}C delivered by the Heavy Ion Research Facility in Lanzhou (HIRFL) [**?**, **?**], located at the Institute of Modern Physics, Chinese Academy of Sciences.

Fig. 1 [Figure 1: see original paper] shows the layout diagram of the neutron source experimental device. In this work, the broad-spectrum neutrons generated by this setup served as source neutrons, interacting with a ^{23} U target sample. The resulting leakage neutron spectrum was then measured. The ^{12}C ions were accelerated to an energy of 80.5 MeV/u and focused onto a natural

tungsten cylinder target (dimensions: 50 mm × 5 mm). The tungsten target was attached to the exit window, which was made of a 3-mm-thick iron plate at the end of the vacuum chamber. The bombardment of the tungsten target by the ^{12}C beam generates a large number of secondary particles, including neutrons, gamma rays, protons, deuterons, tritons, and ^{3}He nuclei. These secondary particles can penetrate through the target medium and the surrounding air, allowing their signals to be detected by the detectors (EJ212 and BC501A).

Fig. 2 [Figure 2: see original paper] shows the layout diagram of the experimental device for the transport of broad-spectrum neutrons in the ^{23}U target (dimensions: 100 mm × 100 mm × 20 mm; thickness along the neutron penetration direction: d = 20 mm; purity: p = 99.9 %; density: = 18.79 g/cm³). In this experiment, the time-of-flight method was employed to measure the leakage neutron spectrum. As shown in the figure, the experimental device is largely consistent with that of the neutron source facility (Fig. 1). The difference is that the plastic scintillation detector (Veto) and the ^{23}U target are placed on the outer side of the vacuum chamber. The Veto is used to remove the effects of charged particles on the leakage neutron spectrum during data analysis. The beam pickup detector (TP, a plastic scintillator detector) positioned within the vacuum chamber is located 67 cm upstream of the tungsten target. During the experiment, the TP served to monitor beam parameters, provide timing information for the beam and count incident particles, while also acting as the stop signal for neutron time-of-flight measurements. Additionally, ^{12}C ions were fully deposited in the primary target (tungsten target), with their range calculated using LISE++ being equal to 1.912 mm. In this experiment, the detection system consisted of an EJ212 plastic scintillator detector (Veto) and a BC501A liquid scintillator detector (N1). This system allowed effective discrimination between charged and neutral particles. After removing signals due to charged particles, the remaining spectrum enabled differentiation between neutron and gamma signals detected by the liquid scintillator via pulse shape discrimination (PSD), as shown in Fig. 3 [Figure 3: see original paper]. The BC501A liquid scintillator detector primarily consists of a cylindrical liquid scintillator crystal, an optical collection system, a photomultiplier tube, and a voltage divider. The liquid scintillator crystal inside the BC501A probe has a diameter and length of 12.7 cm each, a maximum emission wavelength of 425 nm, and an emission decay time of approximately 3.2 ns. The EJ212 plastic scintillation detector is a rectangular cuboid with dimensions of 12.7 cm × 12.7 cm × 5 mm and has a square cross-section facing the incident beam direction. Its distinguishing characteristic is a very low probability of interaction with neutral particles, while it exhibits high sensitivity to charged particles.

B. Electronics and Data Acquisition

Fig. 4 [Figure 4: see original paper] shows a schematic diagram of the electronics and data acquisition system employed in the neutron transport experiment for ^{23}U. As shown in the figure, the signal from the beam pickup detector (TP

or BM) located in the target chamber is divided into four channels. The signal from Channel 1 is fed into a Charge-Integrating Digital Converter (QDC) to record beam energy information. The signal from Channel 2 is processed by a Constant Fraction Discriminator (CFD) and then fed into a Time-to-Digital Converter (TDC) to serve as the stop signal for neutron time-of-flight measurement. The signal from Channel 3 passes through the CFD and enters the Scaler. The signal from Channel 4 is processed by the CFD and then directed to a Logic Coincidence Unit (Coin), where it is used in coincidence with the output signal from the neutron detector to identify true nuclear reaction events. This coincident signal is subsequently sent to the Scaler and also serves as the electronics trigger signal (Trigger) for the entire experiment. Signals from the plastic scintillator detectors (Veto1 and Veto2) are directly input to the QDC for removing charged particle events during data analysis. The output signal from the liquid scintillator detector (BC501A) is split into three channels as illustrated: The Channel 1 signal is processed by the CFD and then fed into the TDC to serve as the start time for neutron time-of-flight measurement; The Channel 2 signal is used for fast/slow component discrimination in the QDC; The Channel 3 signal passes through the CFD and is then input to the coincidence unit for coincidence measurement with the BM signal.

C. Monte Carlo Calculation

GEANT4 is a versatile Monte Carlo software toolkit for the calculation of the passage of particles through matter. It is widely used across various fields, including high-energy physics, accelerator physics, astrophysics and space science, medical physics, and radiation protection. GEANT4 provides a variety of physical models describing the interaction between particles and nuclei, along with comprehensive lists of associated models for users to utilize, such as BERT (Bertini intranuclear cascade model), BIC (Binary cascade model), and INCL++ (Intranuclear cascade Liège). BERT integrates the Bertini intranuclear cascade model, pre-equilibrium model, fission model, and evaporation model. It is capable of simulating nuclear reactions induced by long-lived hadrons and gamma rays with energies up to 10 GeV. BIC simulates the cascade transport process of primary and secondary particles within the nucleus. It only considers the two-body interactions between primary or secondary particles and individual nucleons within the nucleus. INCL++ is applicable for simulating particle bombardment of target nuclei heavier than deuterium within the energy range of 1 MeV/u to 20 GeV/u. However, its applicability to light and unstable nuclei has not yet been comprehensively validated.

In order to verify the reliability of GEANT4 in the transport calculation process, this study employs GEANT4 to simulate the experimental process in combination with the INCL++, BIC, and BERT physical models and the ENDF/B-VIII, JEFF-3.3, and JENDL-4.0 evaluation databases. The resulting leakage neutron spectra are obtained under the same experimental conditions.

III. DATA ANALYZING PROCEDURE

A. Neutron Energy Calculation

The time-of-flight (TOF) method for measuring neutron energy spectra is based on the principle that neutrons with different energies require different amounts of time to travel the same distance. It is a measurement technique that calculates neutron energy based on the known flight distance and corresponding travel time. In this experiment, the flight distance L is defined as the distance between the center of the target sample and the geometric center of the detector.

It should be noted that since the beam pickup detector is positioned at a certain distance upstream from the target, the time difference between the trigger signal and the stop signal cannot be directly used as the neutron time of flight. The actual neutron time of flight is obtained by taking the time difference between the neutron peak and the gamma peak in the TOF spectrum, plus the flight time of the gamma rays from the target center to the detector center. The neutron energy can then be derived from the TOF spectrum using the following Eq. (1):

$$E = (\sqrt{1 - (\frac{L}{\Delta Tc + L})^2}^{-1} - 1)m_n c^2$$

where E is the energy of the neutron, L is the flight distance from the center of the target to the geometric center of the detector, ΔT is the difference of flight times between the prompt gamma ray and the neutron, c is the velocity of light in a vacuum, and m_n is the rest mass of the neutron.

B. Energy Calibration and Neutron-Detection Efficiency

The energy calibration results of the neutron detector directly affect the accuracy and reliability of the leakage neutron spectrum. Therefore, the energy calibration of the detector is of vital importance in neutron transport experiments. Based on the principle that the light output of low-energy electrons in the liquid scintillator detector is linearly related to the electron energy, standard gamma-ray sources (^{13}Cs, Co, and ^{22}Na) were used to calibrate the energy of the BC501A (N1) detector. The energy calibration results are shown in Fig. 5 [Figure 5: see original paper].

The detection efficiency ( ) is defined as the ratio of the number of detected particles to the number of particles incident on the detector. Due to the limitations imposed by detector type, geometric dimensions, energy threshold, the type and energy of the incident particles, as well as the dead time of the data acquisition system, not all neutrons entering the detector can be recorded. To obtain a more accurate neutron leakage spectrum, the neutron detection efficiency must therefore be determined. In this experiment, the SCINFUL-QMD

[**?**] simulation program was used to calculate the detection efficiency of the liquid scintillator detector under different detection thresholds. When the neutron energy is below 80 MeV, the software employs the SCINFUL model for calculations; for energies above 80 MeV but below 3 GeV, it utilizes the Quantum Molecular Dynamics (QMD) model and the Statistical Decay Model (SDM) for calculations. In previous work, the software has been benchmarked and applied to neutron transport experiments. The result is shown in Fig. 6 [Figure 6: see original paper].

C. Uncertainty and Energy Resolution

The uncertainty of the experimental data consists of statistical and systematic errors. The statistical error was less than 5% at energies below 20 MeV and increased to 35% at higher energies from the neutron counts. The systematic error arises mainly from uncertainties in the detection efficiency calculation, being less than 10% for incident neutron energies in the range of 0.1–80 MeV and approximately 15% for higher-energy neutrons. The energy resolution can be expressed by the following Eq. (2):

$$\frac{\Delta E}{E} = \frac{\Upsilon(\Upsilon+1)}{2}\frac{\Delta t}{\Delta T}$$

where Υ is the Lorentz factor, ΔT is the difference in flight times between the prompt gamma ray and the neutron. The time resolution Δt was estimated to be 1.5 ns, which is obtained from the Full Width at Half Maximum (FWHM) of the prompt gamma-ray peak.

IV. RESULTS AND DISCUSSION

In this experiment, broad-spectrum neutrons emitted from a 5 mm-thick tungsten target bombarded by 80.5 MeV/u ^{12}C ions were measured using the time-of-flight method. Subsequently, the leakage neutron spectrum at 0° was measured from the interaction of these broad-spectrum neutrons with a ^{23}U target. Parallel calculations were performed using GEANT4 with the INCL++, BIC, and BERT physics models, combined with the ENDF/B-VIII, JEFF-3.3, and JENDL-4.0 evaluated databases. The leakage neutron spectrum under identical experimental conditions was thereby obtained through these calculations.

Fig. 7 [Figure 7: see original paper] compares the leakage neutron spectrum of the ^{23}U target with the source neutron spectrum (i.e., broad-spectrum neutrons generated by ^{12}C bombardment of the W target). In the figure, the blue solid squares represent the source neutron spectrum. The red solid circles show the leakage neutron spectrum at 0° after the source neutrons transported through the ^{23}U target. The results indicate that the 20 mm-thick ^{23}U target affects neutrons across the entire studied energy range, with a more pronounced influence observed for neutrons below 50 MeV.

Fig. 8 [Figure 8: see original paper] shows a comparison between experimental data and GEANT4 calculation results for the leakage neutron spectrum of the ^{23}U target at the 0° direction. The solid black dots represent the experimental results obtained in this study, while lines in other colors (red, green, and blue) correspond to different physics models (INCL++, BIC, and BERT). The solid lines, dashed lines, and dash-dotted lines respectively represent different evaluated databases (ENDF/B-VIII, JEFF-3.3, and JENDL-4.0). For neutron transport in the ^{23}U target, the calculation results from the INCL++, BIC, and BERT models exhibit similarity in the energy range above 20 MeV. Overall, the calculation results from the three models reproduce the experimental data well. Within the energy range above 80 MeV, the errors of the calculation results of the three models have all increased.

Since GEANT4 utilizes evaluated databases for calculations in the energy range below 20 MeV, a separate comparison between experimental measurements and calculation results for the leakage neutron spectrum below 20 MeV is presented in Fig. 9 [Figure 9: see original paper] to highlight differences among the databases. Below 20 MeV, the calculation results of the three models calling the ENDF/B-VIII, JEFF-3.3, and JENDL-4.0 databases are slightly different. These differences are primarily observed below 10 MeV, where the BERT model yields higher results than INCL++ and BIC. Additionally, in this low-energy range (below 10 MeV), all the calculation results collectively overestimate the experimental data. Above 10 MeV, the results from all three models show good agreement.

V. CONCLUSIONS

The experiment described in this paper was carried out at the Radioactive Ion Beam Line in Lanzhou (RIBLL). The time-of-flight (TOF) method was employed to measure the leakage neutron spectrum at 0° generated by the interaction of broad-spectrum neutrons (high-energy white-light neutrons) with a ^{23}U target. Additionally, calculations were performed using GEANT4 combined with the INCL++, BIC, and BERT physics models, as well as the evaluated nuclear data libraries ENDF/B-VIII, JEFF-3.3, and JENDL-4.0. Under the same experimental conditions, the theoretical results for the leaked neutron spectrum were obtained. The results indicate that the calculations using the three physical models combined with different evaluated nuclear data libraries all accurately reproduced the experimental data. However, slight overestimations were observed below 10 MeV when these models were paired with different data libraries. Overall, GEANT4 demonstrates reliable calculations for the transport process of high-energy neutrons in ^{23}U targets.

This work fills a gap in the studies of high-energy neutron transport for the ^{23}U target. The reliable transport experimental data provide valuable reference information for the design and construction of ADS projects. Additionally, the study examines the reliability of the evaluated databases and theoretical models in the GEANT4 program, and contributes to model improvement and the

refinement of evaluated databases, which is of significant importance.

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