Measurement and calculation of neutron transmission spectra through 238U irradiated by a broad-spectrum neutron field

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Abstract

The neutron transmission spectrum through a high-purity 238U slab (dimensions: 100 mm × 100 mm × 20 mm) irradiated by a broad-spectrum neutron field was measured at 0 ∘ using the time-of-flight (TOF) method. Theexperiment 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. GEANT4 calculations were performed under the same experimental conditions by combining the INCL++, BIC, and BERT physics models with the evaluated nuclear data libraries ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0. The calculations reproduce the measured spectrum reasonably well over most of the investigated energy range; however, they overestimate the data below 10 MeV and tend to underestimate the measured yield above 70 MeV. The present results provide benchmark information for validating neutron-transport simulations relevant to accelerator-driven systems.

Full Text

Preamble

Measurement and calculation of neutron transmission spectra through U irradiated by a broad-spectrum neutron field Hui Sun Xin Zhang, Zhi-Qiang Chen 1, 2, 3 Rui Han, 1, 2, 3, Bo Yang Shakhboz Khasanov Pei-Yan Zhang Guo-Yu Tian, Bing-Yan Liu, Fu-Dong Shi, Ze-Kun Zhang Qin Li, and Peng Luo 1, 2, 3 1 Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China Gansu Isotope Laboratory, Lanzhou 730300, China School of Nuclear Science and Technology, University of Chinese Academy of Sciences, Beijing 100049, China Samarkand State University, Samarkand 140104, Uzbekistan The neutron transmission spectrum through a high-purity U slab (dimensions: 100 mm

100 mm mm) irradiated by a broad-spectrum neutron field was measured at using the time-of-flight (TOF) 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 C ions. GEANT4 calculations were performed under the same experimental conditions by combining the INCL++, BIC, and BERT physics models with the evaluated nuclear data libraries ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0. The calculations reproduce the measured spectrum reasonably well over most of the investigated energy range; however, they overestimate the data below 10 MeV and tend to underestimate the measured yield above 70 MeV. The present results provide benchmark information for validating neutron-transport simulations relevant to accelerator-driven systems.

Keywords

Neutron transmission spectrum, U, Broad-spectrum neutrons, GEANT4

INTRODUCTION

An accelerator-driven system (ADS) couples a subcritical reactor core to an external accelerator-driven spallation neu- tron source [ ]. Such systems are attractive because they can support transmutation of long-lived actinides and fission products, improve fuel-resource utilization, and benefit from the inherent safety margin associated with subcritical oper- ation [ ]. Consequently, ADS has become an important research topic in nuclear energy [ ]. Its continued de- velopment relies strongly on accurate nuclear data and val- idated transport methodologies [ ]. In ADS, the sec- ondary neutrons generated by beam-induced spallation re- actions are characterized by high intensity and a broad en- ergy distribution. These neutrons not only serve as the pri- mary driver for spent fuel transmutation, but the transmit- ted neutrons also constitute an important component of the radiation source term in shielding design. Therefore, inves- tigating neutron transport processes in relevant materials is crucial for the design of core structure, target-core coupling, and radiation shielding. Monte Carlo codes based on estab- lished physical models and evaluated nuclear data libraries are fundamental tools for conducting neutron transport sim- ulations. Accordingly, the optimization of subcritical-system parameters, including geometric configuration and material selection, requires reliable transport calculations over a wide neutron-energy range.

U is a key nuclide in ADS and advanced fuel-cycle systems, and reliable neutron-transport data for U are re- quired for both neutronic design and safety assessment [This work was supported by the National Natural Science Foundation of China (Nos. U1832205, 12005265 and 12305150).]. However, appreciable discrepancies remain among eval- uated neutron data for U below 20 MeV, while bench- mark information at intermediate and high energies is still limited [ ]. Under broad-spectrum neutron irradiation, U undergoes elastic and inelastic scattering, radiative cap- ture, fission, and secondary-particle production; these pro- cesses strongly affect the neutron field in the target region and are therefore directly relevant to ADS analyses [After neutron capture, U can breed fissile Pu through the decay chain Accordingly, reliable experimental measurements and calcu- lations of neutron transport through U are required.

Benchmark measurements of neutron transmission through U under broad-spectrum irradiation provide a direct test of Monte Carlo transport calculations relevant to ADS. Such data are particularly valuable for assessing the combined per- formance of reaction models and evaluated libraries over a wide energy range. The present work therefore helps to re- duce the lack of benchmark information for neutron transport through U at intermediate and high energies.

In this work, broad-spectrum source neutrons produced in C + W reaction were used to irradiate the U sam- ple, and the transmission spectrum was measured by the TOF method. In parallel, the experiment was simulated with GEANT4 [ ] by combining the INCL++ [ ], BIC [and BERT [ ] physics models with the ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0 evaluated nuclear data libraries.

Comparison between measurement and calculation was then used to assess the reliability and limitations of the adopted transport models.

U sample

METHODS

Experimental methodology The experiment was carried out at the Radioactive Ion Beam Line in Lanzhou (RIBLL) [ ], using an 80.5 MeV/u C primary beam.

The beam was delivered in continuous-wave mode by the Heavy Ion Research Facility in Lanzhou (HIRFL) [ ] at the Institute of Modern Physics, Chinese Academy of Sciences.

Figure shows the layout of the neutron source experimen- tal device. In this work, the broad-spectrum neutrons gen- erated by this setup served as the incident neutrons for the U target sample. The transmitted neutron spectrum was subsequently measured. The C ions were accelerated to an energy of 80.5 MeV/u and focused onto a natural tung- sten cylindrical target (diameter 50 mm, thickness 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 vac- uum chamber. The bombardment of the tungsten target by C beam generated a large number of secondary particles, including neutrons, gamma rays, protons, deuterons, tritons, and He nuclei. After traversing the target region and the surrounding air, part of this mixed secondary-particle field reached the EJ212 and BC501A detectors.

U target (dimensions: 100 mm 100 mm 20 mm; thickness along the neutron penetration direction: d = 20 mm; purity: p = 99.9 %; den- sity: = 18.79 g/cm). In this experiment, the time-of-flight method was employed to measure the transmission neutron spectrum. As shown in the figure, the experimental device was largely identical to that used for the neutron-source mea- surement (Figure). The difference is that the plastic scintil- lator detector (Veto) and the U sample were placed outside of the vacuum chamber. The Veto was used to reject charged- particle events 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 beam-timing information and incident- particle counts, and act as the stop signal for the neutron TOF measurement. Additionally, C ions were fully stopped in the primary target (tungsten target), with their range calcu- lated using LISE++ being equal to 1.912 mm. In this experi- ment, the detection system consisted of an EJ212 plastic scin- tillator detector (Veto) and a BC501A liquid scintillator de- tector (N1). This system enabled charged-particle rejection; after removal of charged-particle events, neutron and gamma signals recorded by the liquid scintillator were separated by pulse shape discrimination (PSD), as shown in Figure . The BC501A liquid scintillator detector consists of a cylindrical liquid-scintillator cell, 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 scintillator detector is a rectangular plate with dimensions of 12.7 cm 12.7 cm 5 mm and has a square cross-section facing the incident beam direction. Its distin- guishing characteristic is a very low probability of interaction with neutral particles, while it exhibits high sensitivity to charged particles.

B. Electronics and data acquisition

238 U. As shown in the figure, the signal from the beam pickup detector (TP, denoted BM in the electronics diagram) located in the target chamber is divided into four

channels. The signal from Channel 1 was fed into a charge- to-digital converter (QDC) to record beam-energy informa- tion. 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 neu- tron 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 co- incidence 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 elec- tronics trigger signal (Trigger) for the entire experiment. Sig- nals 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 was processed by the CFD and then fed into the TDC as the start signal for neutron TOF measurement; the Channel 2 signal was integrated over short and long gates in the QDC for PSD; and the Channel 3 signal passed through the CFD before entering the coincidence unit for coincidence measurement with the BM signal.

Monte Carlo calculations 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 pro-

vides a variety of physics models describing the interac- tion 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++ (Liège intranuclear cascade model).

BERT combines the Bertini intranuclear cascade model, pre- equilibrium model, fission model, and evaporation model. It is capable of simulating nuclear reactions induced by hadrons and gamma rays with energies up to 10 GeV. BIC simulates the cascade transport process of primary and secondary par- ticles within the nucleus. It only considers the two-body in- teractions between primary or secondary particles and indi- vidual 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.

To benchmark the reliability of GEANT4 for the present transport problem, version 10.7.4 was used to simulate the ex- periment by combining the INCL++, BIC, and BERT physics models with the ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0 evaluated databases. The transmission neutron spectra were then calculated under the same experimental conditions as in the measurement.

DATA ANALYSIS PROCEDURE 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, and it is now widely used in neutron experiments [40 -43]. 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 posi-

tioned at a certain distance upstream from the target, the time difference between the trigger signal and the stop signal can- not be directly used as the neutron time of flight. The actual neutron time of flight is obtained by taking the time differ- ence 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. Figure displays the time-of-flight spectra of neutrons and gamma rays, with the charged-particle background removed. The neutron energy can then be derived from the TOF spectrum using Eq. (

E = (1

where is the energy of the neutron, is the flight dis- tance from the center of the target to the geometric center of the detector, is the difference in flight time between the prompt gamma ray and the neutron, is the velocity of light in a vacuum, and is the neutron rest mass.

Energy calibration and neutron-detection efficiency The energy calibration of the neutron detector directly af- fects the accuracy and reliability of the transmission spec- trum and is therefore essential in neutron-transport experi- ments. 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 Co, and Na) were used to calibrate the energy of the BC501A (N1) detector. The energy calibration results are shown in Fig.

The detection efficiency ( ) is defined as the ratio of the number of detected particles to the number of particles inci- dent on the detector. Due to the limitations imposed by de- tector 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 transmission 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 be- low 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 previ- ous work, the software has been benchmarked and applied to neutron transport experiments. The result is shown in Fig.

Experimental background Background neutron measurements were conducted to evaluate the background contribution of scattered neutrons in the experimental hall. Figure shows the experimental setup

for measuring scattered background neutrons. A shadow bar made of iron, measuring 15 cm 15 cm in cross-section and 100 cm in length, was placed along the flight path from the target to the liquid scintillator detector. This setup blocked almost all direct neutrons, allowing only the scattered com- ponents to reach the neutron detector. Figure displays the time-of-flight spectrum of the background neutrons.

Uncertainty and energy resolution The uncertainty of the experimental data consists of statis- tical and systematic components. The statistical uncertainty was less than 5 % below 20 MeV and increased to about 35 % at the highest energies because of limited neutron statistics.

The dominant systematic component arose from the uncer- tainty in the calculated detection efficiency; it was estimated to be less than 10 % for incident neutron energies in the range 0.1–80 MeV and approximately 15 % for higher-energy neu- trons. The energy resolution can be expressed by the follow- ing Eq. (where is the Lorentz factor, is the flight distance from the center of the target to the geometric center of the detector, is the uncertainty in the flight distance and was less than 1 cm, is the difference in flight time between the prompt gamma ray and the neutron, and the time resolution estimated to be 1.5 ns from the full width at half maximum (FWHM) of the prompt gamma-ray peak shown in Fig.

RESULTS AND DISCUSSION In this experiment, broad-spectrum neutrons emitted from a 5 mm-thick tungsten target bombarded by 80.5 MeV/u ions were characterized by the TOF method. The transmis- sion neutron spectrum at was then measured for the sample. Parallel calculations were performed with GEANT4 using the INCL++, BIC, and BERT physics models in com- bination with the ENDF/B-VIII.0, JEFF-3.3, and JENDL- 4.0 evaluated databases, and the corresponding transmission spectra were obtained under the same experimental condi- tions.

Figure compares the transmission neutron spectrum of U target with the source neutron spectrum (i.e., the broad-spectrum neutrons generated by C bombardment of the W target). In the figure, the blue solid squares represent the source neutron spectrum. The red solid circles show the transmission neutron spectrum at 0° after the source neutrons passed through the U target. The results indicate that the

U target at 20 mm-thick U target modifies the neutron field over the entire investigated energy range, with a more pronounced ef- fect below 50 MeV.

Figure shows a comparison between experimental data and GEANT4 calculation results for the transmission neu- tron spectrum of the U target at the 0° direction. 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.0, JEFF-3.3, and JENDL-4.0). As shown in , the GEANT4 results reproduce the measured spec- trum reasonably well over most of the investigated energy range.

Because the transport calculation below 20 MeV is mainly controlled by the selected evaluated libraries in GEANT4, a separate comparison for this energy region is presented in Fig. to highlight the differences among the databases.

Figure presents the ratio between the calculated and experimental values. As shown in the figure, in the 20–70 MeV energy range the results from all three models agree U target

U target at below 20 reasonably well with the experimental data, with discrepancies generally within 15%. Above 70 MeV, the deviations in- crease and all model combinations clearly underestimate the measured yield. Overall, for broad-spectrum neutron trans- port through U, GEANT4 combined with the three physics models provides reasonable agreement above 20 MeV, but the description deteriorates in the highest-energy region, in- dicating that the high-energy reaction treatment still requires improvement. A combined analysis of Figs.

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Conf. (2022). A. Rummana, R. J. Barlow, S. M. Saad, et al ., Cal- culations of neutron fluxes and isotope conversion rates in a thorium-fuelled MYRRHA reactor, using GEANT4 ther shows that, below 20 MeV, differences among ENDF/B- VIII.0, JEFF-3.3, and JENDL-4.0 are small for a given cas- cade model. However, below 10 MeV all calculations over- estimate the experimental data, and the BERT model gives slightly higher values than INCL++ and BIC in this low- energy region.

Above 10 MeV, the agreement among the three models becomes noticeably better.

CONCLUSIONS

The experiment reported here was carried out at the Ra- dioactive Ion Beam Line in Lanzhou (RIBLL). The TOF method was used to measure the neutron transmission spec- trum at for a U slab irradiated by broad-spectrum neu- trons generated in 80.5 MeV/u C + W reactions. Corre- sponding calculations were performed with GEANT4 using the INCL++, BIC, and BERT physics models together with the evaluated nuclear data libraries ENDF/B-VIII.0, JEFF- 3.3, and JENDL-4.0. Under the same experimental condi- tions, the corresponding calculated transmission spectra were obtained.

The calculations reproduce the measured spectrum reason- ably well over most of the investigated energy range. In par- ticular, the discrepancy is generally within 15% in the range 20–70 MeV. Below 10 MeV, all model-library combinations overestimate the measured yield, whereas above 70 MeV they tend to underestimate it. These deviations indicate that further improvement of the low- and high-energy reaction description remains desirable.

The present measurement provides benchmark information for validating neutron-transport calculations in U relevant to ADS target, shielding, and fuel-cycle studies. These data are also useful for assessing the consistency of evaluated li- braries and for guiding future refinement of transport models.

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Author contributions All authors contributed to the study conception and design. Material preparation, data collection, and data analysis were performed by Hui Sun, Xin Zhang, and Rui Han. Theoretical calculations and interpretation were performed by Hui Sun and Zhi-Qiang Chen.

The project was led and supervised by Zhi-Qiang Chen, Rui Han, and Hui Sun. The first draft of the manuscript was written by Hui Sun, and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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