



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

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Abstract

The neutron transmission spectrum through a high-purity ^{238}U 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. 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 ^{12}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 · ^{238}U · Broad-spectrum neutrons · GEANT4

1 Introduction

An accelerator-driven system (ADS) couples a subcritical reactor core to an external accelerator-driven spallation neutron source [1–3]. 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 operation [4, 5]. Consequently, ADS has become an important research topic in nuclear energy [6–12]. Its continued development relies strongly on accurate nuclear data and validated transport methodologies [13–17]. In ADS, the secondary neutrons generated by beam-induced spallation

reactions are characterized by high intensity and a broad energy distribution. These neutrons not only serve as the primary driver for spent fuel transmutation, but the transmitted neutrons also constitute an important component of the radiation source term in shielding design. Therefore, investigating 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 established physical models and evaluated nuclear data libraries are fundamental tools for conducting neutron-transport simulations. Accordingly, the optimization of subcritical-system parameters, including geometric configuration and material selection, requires reliable transport calculations over a wide neutron energy range.

^{238}U is a key nuclide in ADS and advanced fuel-cycle systems, and reliable neutron-transport data for ^{238}U are required for both neutronic design and safety assessment [17–20]. However, appreciable discrepancies remain among evaluated neutron data for ^{238}U below 20 MeV, while benchmark information at intermediate and high energies is still limited [21–26]. Under broad-spectrum neutron irradiation, ^{238}U undergoes elastic and inelastic scattering, radiative capture, fission, and secondary-particle production; these

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processes strongly affect the neutron field in the target region and are therefore directly relevant to ADS analyses [17, 20]. After neutron capture, ^{238}U can breed fissile ^{239}Pu through the decay chain $^{238}\text{U}(n,\gamma)^{239}\text{U} \rightarrow ^{239}\text{Np} \rightarrow ^{239}\text{Pu}$ [18, 27]. Accordingly, reliable experimental measurements and calculations of neutron transport through ^{238}U are required.

Benchmark measurements of neutron transmission through ^{238}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 performance of reaction models and evaluated libraries over a wide energy range. The present work therefore helps to reduce the lack of benchmark information for neutron transport through ^{238}U at intermediate and high energies.

In this work, broad-spectrum source neutrons produced in the $^{12}\text{C} + \text{W}$ reaction were used to irradiate the ^{238}U sample, and the transmission spectrum was measured by the TOF method. In parallel, the experiment was simulated with GEANT4 [28] by combining the INCL++ [29], BIC [30], and BERT [31, 32] physics models with the ENDF/B-VIII.0 [33], JEFF-3.3 [34], and JENDL-4.0 [35] evaluated nuclear data libraries. Comparison between measurement and calculation was then used to assess the reliability and limitations of the adopted transport models.

2 Methods

2.1 Experimental methodology

The experiment was carried out at the Radioactive Ion Beam Line in Lanzhou (RIBLL) [36, 37], using an 80.5 MeV/u ^{12}C primary beam. The beam was delivered in continuous-wave mode by the Heavy Ion Research Facility in Lanzhou (HIRFL) [38, 39] at the Institute of Modern Physics, Chinese Academy of Sciences.

Figure 1 shows the layout of the neutron source experimental device. In this work, the broad-spectrum neutrons generated by this setup served as the incident neutrons for the ^{238}U target sample. The transmitted neutron spectrum was subsequently measured. The ^{12}C ions were accelerated to an energy of 80.5 MeV/u and focused onto a natural tungsten 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 vacuum chamber. The bombardment of the tungsten target by the ^{12}C beam generated a large number of secondary particles, including neutrons, gamma rays, protons, deuterons, tritons, and ^3He nuclei. After traversing the target region and

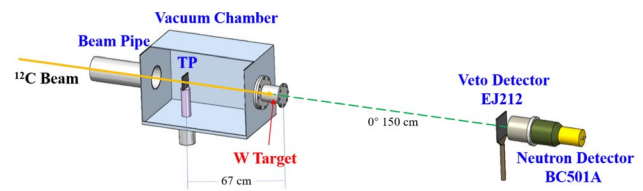


Fig. 1 (Color online) Schematic layout of the neutron source experimental setup

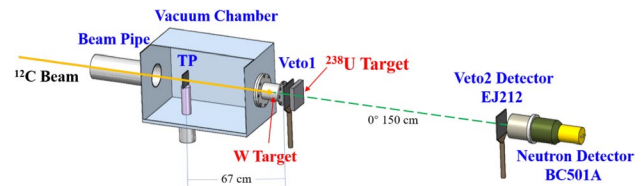


Fig. 2 (Color online) Schematic layout of the experimental setup for neutron transport through the ^{238}U sample

the surrounding air, part of this mixed secondary-particle field reached the EJ212 and BC501A detectors.

Figure 2 shows the experimental setup for the transport of broad-spectrum neutrons in the ^{238}U target (dimensions: 100 mm $\times$ 100 mm $\times$ 20 mm; thickness along the neutron penetration direction: $d = 20$ mm; purity: $p = 99.9\%$; density: $\rho = 18.79$ g/cm³). 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 measurement (Fig. 1). The difference is that the plastic scintillator detector (Veto) and the ^{238}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, ^{12}C ions were fully stopped 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 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 Fig. 3. 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

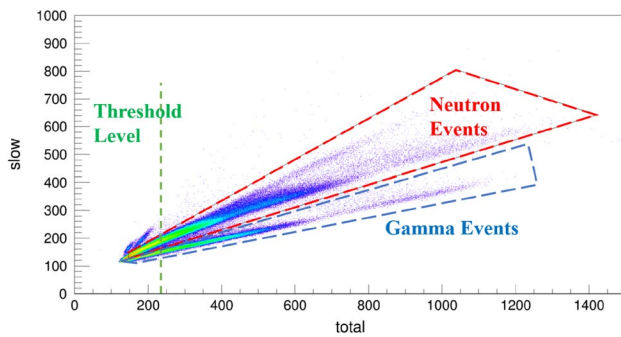


Fig. 3 (Color online) QDC output spectrum with total and slow gates

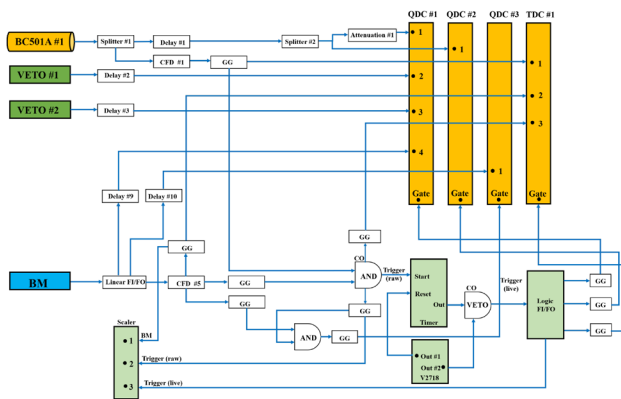


Fig. 4 (Color online) Schematic of the electronics and data acquisition system

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 $\times$ 12.7 cm $\times$ 0.5 cm 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.

2.2 Electronics and data acquisition

Figure 4 shows a schematic diagram of the electronics and data acquisition system employed in the neutron-transport experiment for ^{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 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 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.

2.3 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 provides a variety of physics 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++ (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 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.

To benchmark the reliability of GEANT4 for the present transport problem, version 10.7.4 was used to simulate the experiment 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.

3 Data analysis procedure

3.1 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 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. Figure 5 displays the time-of-flight spectra of neutrons and gamma

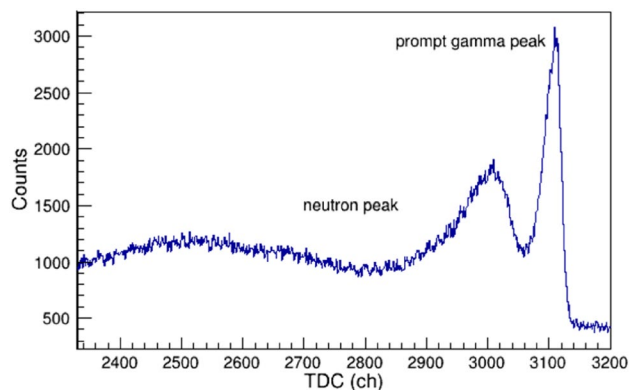


Fig. 5 Time-of-flight (TOF) spectrum of neutrons and gamma rays measured at 0°

rays, with the charged-particle background removed. The neutron energy can then be derived from the TOF spectrum using Eq. (1):

$$E = \left(\frac{1}{\sqrt{1 - \left(\frac{L}{\Delta T c + L} \right)^2}} - 1 \right) m_n c^2, \quad (1)$$

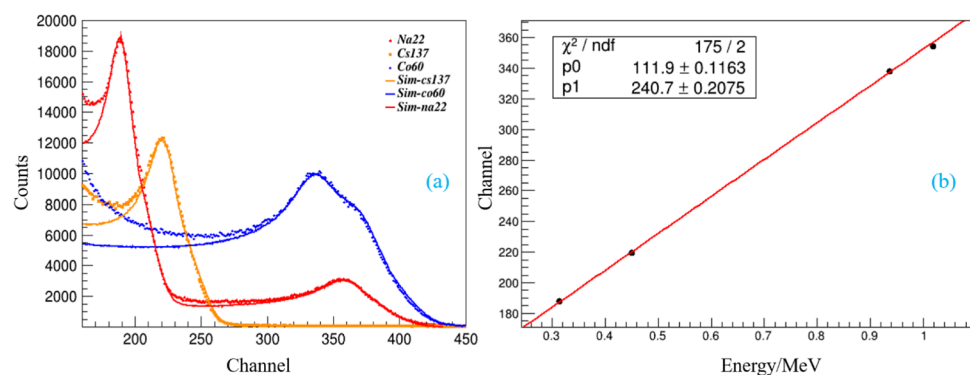
where E is the energy of the neutron, L is the flight distance from the center of the target to the geometric center of the detector (In this experiment $L = 150$ cm.), ΔT is the difference in flight time between the prompt gamma ray and the neutron, c is the velocity of light in a vacuum, and m_n is the neutron rest mass.

3.2 Energy calibration and neutron-detection efficiency

The energy calibration of the neutron detector directly affects the accuracy and reliability of the transmission spectrum and is therefore essential 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 (^{137}Cs , ^{60}Co , and ^{22}Na) were used to calibrate the energy of the BC501A (N1) detector. The energy calibration results are shown in Fig. 6.

The detection efficiency (η) 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 transmission spectrum, the neutron-detection efficiency must therefore be determined. In this experiment, the SCINFUL-QMD [44, 45] simulation program was used to calculate the detection efficiency of the liquid scintillator detector under different detection thresholds. When the

Fig. 6 (Color online) QDC spectra for the standard sources **a** and the energy calibration result **b** for the BC501A detector



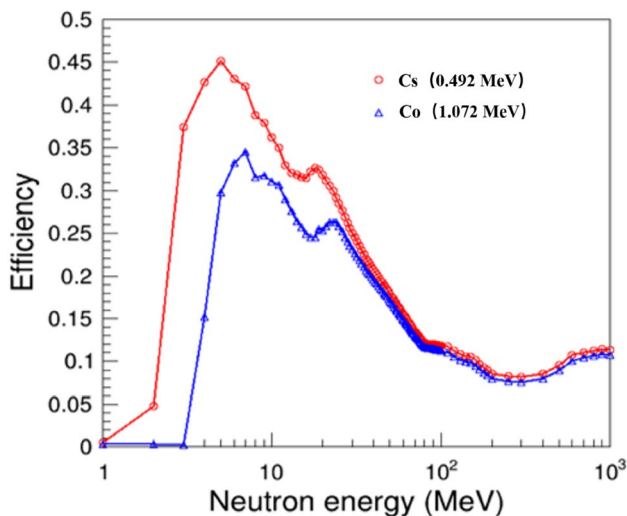


Fig. 7 (Color online) Calculated neutron-detection efficiency of the BC501A detector

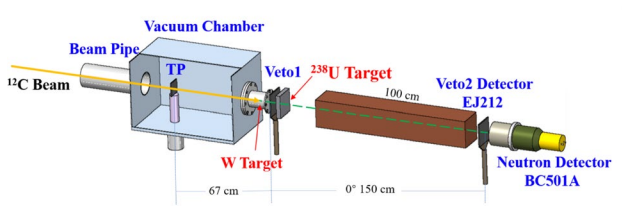


Fig. 8 (Color online) Schematic layout for the background-neutron measurement setup

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

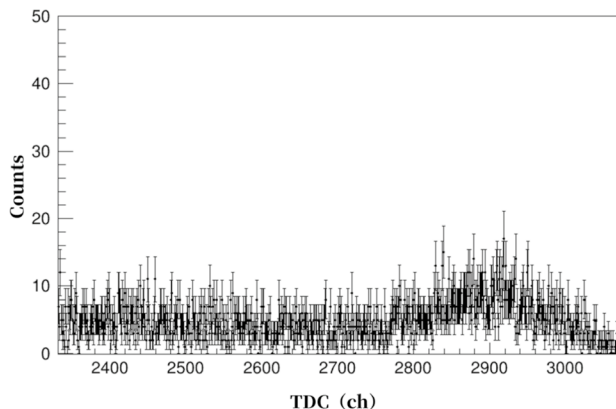


Fig. 9 Time-of-flight (TOF) spectrum of the background neutrons

been benchmarked and applied to neutron-transport experiments. The result is shown in Fig. 7.

3.3 Experimental background

Background neutron measurements were conducted to evaluate the background contribution of scattered neutrons in the experimental hall. Figure 8 shows the experimental setup for measuring scattered background neutrons. A shadow bar made of iron, measuring 15 cm $\times$ 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 components to reach the neutron detector. Figure 9 displays the time-of-flight spectrum of the background neutrons.

3.4 Uncertainty and energy resolution

The uncertainty of the experimental data consists of statistical 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 uncertainty 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 neutrons. The energy resolution can be expressed by the following Eq. (2):

$$\frac{\Delta E}{E} = Y(Y+1) \sqrt{\left(\frac{\Delta L}{L}\right)^2 + \left(\frac{\Delta t}{\Delta T}\right)^2} \quad (2)$$

where Y is the Lorentz factor, L is the flight distance from the center of the target to the geometric center of the detector, ΔL is the uncertainty in the flight distance and was less than 1 cm, ΔT is the difference in flight time between the prompt gamma ray and the neutron, and the time resolution Δt is estimated to be 1.5 ns from the full width at half maximum (FWHM) of the prompt gamma-ray peak shown in Fig. 5.

4 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 characterized by the TOF method. The transmission neutron spectrum at 0° was then measured for the ^{238}U sample. Parallel calculations were performed with GEANT4 using the INCL++, BIC, and BERT physics models in combination with the ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0 evaluated databases, and the corresponding

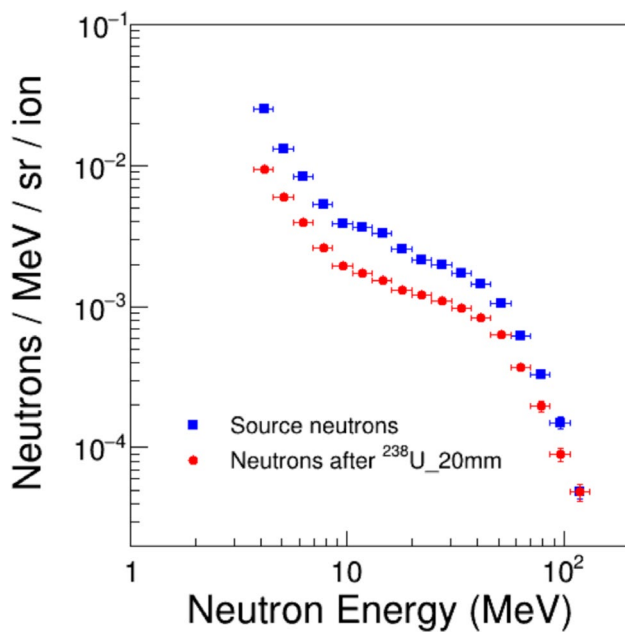


Fig. 10 (Color online) Comparison of the source neutron spectrum and the transmission neutron spectrum for the ^{238}U target

transmission spectra were obtained under the same experimental conditions.

Figure 10 compares the transmission neutron spectrum of the ^{238}U target with the source neutron spectrum (i.e., the 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 transmission neutron spectrum at 0° after the source neutrons passed through the ^{238}U target. The results indicate that the 20-mm-thick ^{238}U target modifies the neutron field over the entire investigated energy range, with a more pronounced effect below 50 MeV.

Figure 11 shows a comparison between experimental data and GEANT4 calculation results for the transmission neutron spectrum of the ^{238}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.0, JEFF-3.3, and JENDL-4.0). As shown in Fig. 11, the GEANT4 results reproduce the measured spectrum 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. 12 to highlight the differences among the databases.

Figure 13 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

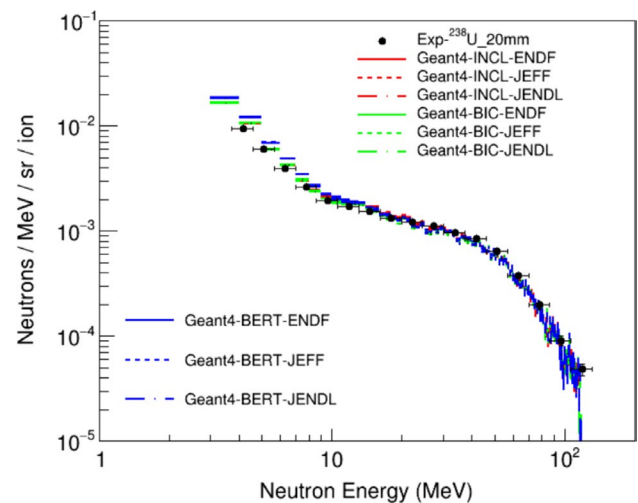


Fig. 11 (Color online) Comparison between experimental data and calculations for the transmission neutron spectrum of the ^{238}U target at 0°

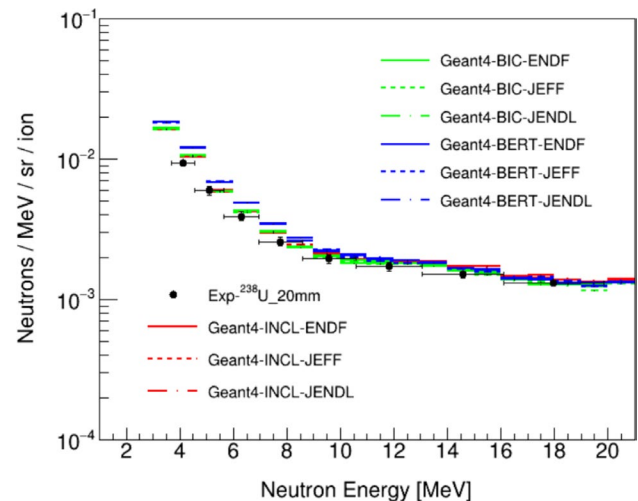


Fig. 12 (Color online) Comparison between experimental data and calculations for the transmission neutron spectrum of the ^{238}U target at 0° below 20 MeV

reasonably well with the experimental data, with discrepancies generally within 15%. Above 70 MeV, the deviations increase and all model combinations clearly underestimate the measured yield. Overall, for broad-spectrum neutron transport through ^{238}U , GEANT4 combined with the three physics models provides reasonable agreement above 20 MeV, but the description deteriorates in the highest energy region, indicating that the high-energy reaction treatment still requires improvement. A combined analysis of Figs. 12 and 13 further shows that, below 20 MeV, differences among ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0 are small for a given cascade model. However, below 10 MeV all calculations overestimate the experimental data, and the BERT

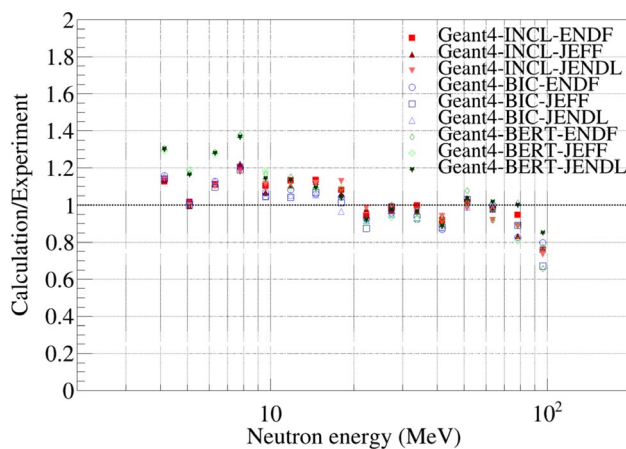


Fig. 13 (Color online) Ratio of calculated to experimental values

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.

5 Conclusion

The experiment reported here was carried out at the Radioactive Ion Beam Line in Lanzhou (RIBLL). The TOF method was used to measure the neutron transmission spectrum at 0° for a ^{238}U slab irradiated by broad-spectrum neutrons generated in $80.5 \text{ MeV/u } ^{12}\text{C} + \text{W}$ reactions. Corresponding 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 conditions, the corresponding calculated transmission spectra were obtained.

The calculations reproduce the measured spectrum reasonably well over most of the investigated energy range. In particular, 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 ^{238}U relevant to ADS target, shielding, and fuel-cycle studies. These data are also useful for assessing the consistency of evaluated libraries 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.

Data availability The data that support the findings of this study are openly available in Science Data Bank at <https://cstr.cn/31253.11.sciencedb.j00186.00833> and <https://www.doi.org/10.57760/sciencedb.j00186.00833>.

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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