Integral shielding experiment on slab zirconium samples using a D–T neutron source

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Abstract

Zirconium (Zr) and its alloys are key structural materials in nuclear reactors due to their low neutron absorption cross section, high-temperature stability, and excellent corrosion resistance. The accuracy and reliability of evaluated nuclear data for Zr isotopes are directly linked to the safety and efficiency of nuclear engineering applications. To provide experimental benchmarks for improving Zr nuclear data, we measured leakage neutron time-of-flight (TOF) spectra for natural Zr samples of three different thicknesses at six scattering angles, using a D–T fusion neutron source in an integral experimental setup. The experimental results were compared with simulated TOF spectra generated using the Monte Carlo N-Particle (MCNP) transport code together with several evaluated nuclear data libraries, including CENDL-3.2, ENDF/B-VIII.0, JEFF-3.3, and JENDL-5. Analysis of the calculated-to-experimental ratios shows that: (1) the CENDL-3.2 library slightly underestimates elastic scattering at small angles but substantially overestimates it at larger angles and in the discrete inelastic scattering regions; (2) the ENDF/B-VIII.0 library significantly underestimates the discrete inelastic scattering regions; (3) the JEFF-3.3 library systematically overestimates the measurements in both the elastic and discrete inelastic scattering regions; and (4) among all libraries considered, JENDL-5 exhibits the best overall agreement with the experimental data. These findings reveal notable inconsistencies in current evaluated nuclear data for Zr isotopes and underscore the need for further refinement to improve their accuracy and reliability.

Shielding Integral Experiment of Slab Zr Samples Based on a D-T Neutron Source

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Zirconium (Zr) and its alloys are critical materials in nuclear reactors because of their low neutron absorption cross-section, high-temperature stability, and excellent corrosion resistance. The accuracy and reliability of evaluated nuclear data for Zr isotopes are directly related to the safety and efficiency of nuclear engineering applications.

To provide experimental data for refining Zr nuclear data, we measured leakage neutron time-of-flight (TOF) spectra for natural Zr samples of three thicknesses at six angles using a D-T fusion neutron source in an integral experimental setup. The experimental results were compared with simulated TOF spectra generated using the Monte Carlo N-Particle Transport Code and nuclear data libraries, including CENDL-3.2, ENDF/B-VIII.0, JEFF-3.3, and JENDL-5. Analysis of the calculated-to-experimental ratios revealed the following: (1) The CENDL-3.2 library slightly underestimated elastic scattering at small angles but significantly overestimated it at larger angles and in discrete inelastic scattering ranges. (2) The ENDF/B-VIII.0 library significantly underestimated the discrete inelastic scattering ranges. (3) The JEFF-3.3 library consistently overestimated the measurements in both elastic and discrete inelastic scattering ranges. (4) The JENDL-5 library demonstrated the best agreement with experimental data among all libraries. These results highlight inconsistencies in existing nuclear data for Zr isotopes and emphasize the necessity for further refinement to enhance their accuracy and reliability.

Keywords: Slab Zr, Leakage neutron spectra, Shielding integral experiment, Evaluated nuclear data

INTRODUCTION

Zirconium (Zr) exhibits excellent nuclear properties, including a low thermal neutron absorption cross-section (1.8×10^{-5} b), good high-temperature stability, and exceptional corrosion resistance [1–5]. Zr alloys can withstand harsh reactor environments while minimizing neutron absorption, thereby enhancing nuclear reaction efficiency. Consequently, Zr alloys are commonly used for fuel cladding and cladding tubes. Additionally, Zr alloys serve as structural materials in other reactor components, such as pressure tubes, active area support structures, and nuclear fuel cores. These components play critical roles in reactor operation and must withstand extreme working conditions. With continuous advances in nuclear power technology and increasing global demand for clean energy, prospects for Zr applications in the nuclear industry are increasingly promising.

Before nuclear data can be effectively utilized, they must undergo integral validation testing to verify their accuracy and reliability. Fusion neutron integral experiments represent a key method for validating nuclear data. By comparing results from integral experimental measurements with Monte Carlo simulations, we can assess discrepancies between various nuclear data libraries. This comparison aids in identifying limitations of each library, thereby providing valuable guidance to nuclear data developers for improving libraries [6–9].

Owing to widespread Zr alloy applications in the nuclear industry, research on Zr-related nuclear data has been ongoing. Although numerous international differential experiments involving Zr have been performed, integral experimental measurements remain relatively scarce due to the high cost of obtaining high-purity, large-sized Zr samples. Previous studies include measurement of leakage neutron spectra for spherical Zr samples irradiated by 14 MeV neutrons at Kyoto University [10], high-energy neutron scattering experiments conducted at Rensselaer Polytechnic Institute [11], and integral experiments using a Cockcroft-Walton accelerator at the China Institute of Atomic Energy (CIAE) [12]. These studies revealed that simulation results based on currently evaluated Zr data still exhibit discrepancies with experimental data, highlighting the necessity for further improvement of Zr isotope evaluation data. Conducting additional shielding integral experiments using Zr samples is essential to provide sufficient high-quality data for refining Zr evaluated data.

In this study, Zr samples were irradiated with 14.5 MeV D-T neutrons, and leakage neutron spectra were measured at six angles using the time-of-flight (TOF) method with an EJ-301 liquid scintillator detector. Monte Carlo simulations were subsequently performed using the Monte Carlo N-Particle Transport Code (MCNP) program [13, 14] to analyze Zr-related data from the CENDL-3.2 [15], ENDF/B-VIII.0 [16], JEFF-3.3 [17], and JENDL-5 [18] libraries. When 14.5 MeV neutrons interact with Zr samples, different reaction channels produce secondary neutrons of varying energies. These reaction channels include elastic scattering, discrete energy-level inelastic scattering, continuous energy-level inelastic scattering, and (n, 2n) reactions. Each channel primarily contributes to different time windows in the TOF spectra. A preliminary evaluation of Zr-related data in different nuclear libraries was conducted by analyzing calculated/experimental (C/E) deviations from various libraries and combining these with cross-section information from each reaction channel.

Compared with earlier integral experiments involving Zr, this study extended leakage neutron spectral measurements to a wider angular range spanning from 47° to 133°, thereby providing a more comprehensive angular dataset. Furthermore, measurements incorporated Zr samples of three different thicknesses, enabling collection of more diverse and detailed experimental data. Additionally, this study validated nuclear data for Zr from the latest release of China' s Evaluated Nuclear Data Library (CENDL-3.2), enhancing the credibility and applicability of the dataset.

II. EXPERIMENTAL SETUP

The experiment was conducted using a 400 kV ns pulsed accelerator at CIAE. A shielding integral experimental setup, developed by the China Nuclear Data Center based on this accelerator, was utilized [19–25]. The experimental system included a T-Ti target, an associated particle detection system, samples, a shielding collimation system, and a monitoring system, all housed in a 20-m-long, 10-m-wide experimental hall with 2-m-thick heavy concrete walls. The main detector was placed in an adjacent hall to ensure that neutrons leaking from the sample traveled more than 7 m before reaching the detector. This arrangement improved neutron energy resolution and minimized background interference.

The beamline was positioned perpendicularly to the collimation line and maintained at an approximate height of 2 m. The experimental setup is shown in Fig. 1 [Figure 1: see original paper].

A. Neutron Source

The 400 kV ns pulsed neutron generator [26] was used to produce pulsed neutrons for the experiment. The generator emits monoenergetic 14.5 MeV fast neutrons through the T(d, n)4He reaction, achieved by bombarding a T-Ti target with a D+ ion beam. The incident D+ ion beam energy was 300 keV, with an average current of 20 µA, pulse width of 2 ns, and pulse frequency of 1.5 MHz. The T-Ti target, with an active zone diameter of 1.6 cm and thickness of 1 mg/cm^2, generated a neutron yield of approximately 2×10^9 n/s.

B. Samples and Measured Angles

Polyethylene was selected as the standard sample to validate the measurement system because of its well-characterized n-p elastic scattering cross-section [27]. The interaction between neutrons and hydrogen involves only the elastic scattering reaction channel (n-p scattering), and the n-p scattering cross-section is internationally recognized as a standard reference. By employing polyethylene, we could effectively validate the experimental system' s reliability, including its data acquisition and processing capabilities, before measuring the natZr samples.

Leakage neutron spectra were measured at six angles to ensure comprehensive data collection. For the 6-cm Zr sample, measurements were conducted at 47°, 61°, 79°, 101°, 119°, and 133°. For the 12-cm and 18-cm samples, the angles were adjusted to 47°, 58°, 73°, 107°, 122°, and 133° to minimize interference with the monitoring system. Measurements were performed at different angles using an electric platform for precise sample positioning, as illustrated in Fig. 2 [Figure 2: see original paper].

Table 1 . Details of samples.

Sample	Size (cm)	Density (g/cm^3)	Major impurity (%)
natZr_1	30 × 30 × 6	-	O(< 0.042); Hf(< 0.01)
natZr_2	30 × 30 × 6	-	Fe(< 0.01); Cr(< 0.01)
natZr_3	30 × 30 × 6	-	Cl(< 0.01)

C. Shield and Collimator

The shielding and collimation system was designed to minimize scattered neutron background and ensure measurement accuracy [28]. It consists of three main components: a shadow bar, pre-collimator, and wall collimator. The shadow bar, positioned between the neutron source and pre-collimator, blocks direct neutron interactions with the pre-collimator material, preventing generation of secondary neutrons.

The wall and pre-collimators have a three-layer structure, as summarized below:

- **First layer (iron):** Reduces fast neutron energy via inelastic scattering.
- **Second layer (polyethylene):** Further reduces neutron energy to the thermal range through elastic scattering.
- **Third layer (lead):** Absorbs gamma rays generated during neutron interactions.

A standard polyethylene sample and three natural Zr (natZr) slabs were used in the experiment. The polyethylene sample dimensions were 30 cm × 30 cm × 6 cm with a density of 0.942 g/cm^3. The Zr sample purity reached 99.5%; detailed information is provided in Table 1.

This system ensures that only neutrons scattered from the sample within the detector’ s solid angle reach the main detector, while background neutrons and gamma rays are effectively suppressed.

D. Detector Systems and Data Acquisition

The experimental setup included an EJ-301 liquid scintillator as the primary detector, two monitor detectors, and an associated particle detector. The EJ-301 detector (diameter: 5.08 cm, thickness: 5.08 cm) measured leakage neutrons emitted from the sample. Two monitor detectors (diameter: 1.27 cm, thickness: 1.27 cm), positioned at 0° and 90° relative to the beam direction, primarily measured TOF spectra of source neutrons. An associated particle detector based on a SiC detector was positioned at 135° to the beam direction to measure alpha particles and protons for neutron yield normalization [29–32].

Neutron yields were calculated using the following relationships:

$$N_n = K_\alpha \times N_\alpha,$$

$$N_n = K_p \times N_p,$$

where N_n is the neutron yield, N_α is the alpha particle count, and N_p is the proton count. K_α and K_p are the corresponding normalization coefficients, derived from the relationship between neutron flux and alpha particle (or proton) flux in the unit solid angle at 135°. These calculations provide critical inputs for normalizing experimental results.

The data-acquisition system employed was GDDAQ [33], which uses Pixie-16 modules for digital signal processing. The key parameters of the Pixie-16 card are listed in Table 2. The system efficiently handled data collection, enabling integration of TOF and pulse shape discrimination (PSD) analyses during post-processing.

Table 2 . Specifications of the Pixie-16 data acquisition module.

Parameter	Specification
Structure	6U PXI/PXIe
Number of channels	-
Sampling frequency	500 MHz
Resolution	12 bit
Maximum waveform capture duration	20 μs
Data transmission rate	1 GB/s

Figure 3 [Figure 3: see original paper] illustrates the TOF measurement system. The pick-up signal generated by the D+ ion beam as it passed through a copper ring inside the tube in front of the target was sent to an acquisition card through a fast pre-amplifier. Signals from the three liquid scintillator detectors and beam pickup signal were processed together to calculate neutron TOF.

E. Experimental Methods and Data Processing

1. Inverse TOF Method The inverse TOF method was adopted to optimize measurement efficiency and reduce system dead time. In this approach, the detector signal served as the start signal and the beam pickup signal served as the stop signal. This method mitigated two major problems:

- It avoided excessive data load on the acquisition system caused by the high-frequency (1.5 MHz) beam pickup signal.

- It reduced system dead time resulting from the low-probability event of neutron detection, which often left start signals without corresponding stop signals.

The resulting inverse TOF spectrum was inverted during data processing to obtain the true TOF spectrum.

2. Data Processing Energy calibration of the EJ-301 detector was performed using standard gamma sources to ensure energy linearity and set measurement thresholds, which were essential for excluding noise signals. Gamma rays were inevitably produced during neutron interactions with matter. The PSD method was employed to distinguish neutron signals from gamma signals by leveraging differences in their pulse shapes.

A baseline correction was applied to each signal to reduce noise. The PSD value was calculated as the ratio of the slow component integral to the total signal integral, enabling effective separation of neutron signals from gamma signals. The impact of PSD on TOF spectra is shown in Fig. 4 [Figure 4: see original paper], where improved neutron spectra are evident after eliminating gamma-ray signals.

During the experiment, source neutrons interacted not only with the sample but also scattered with air and surrounding structural materials. Because the measured leakage neutron spectrum specifically represents neutrons emitted from the sample after interactions with source neutrons, contributions from neutrons scattered by air and other materials had to be eliminated. To achieve this, we performed background measurements with the D-T neutron source present but without the sample. The resulting background spectrum was subsequently compared with the effect spectrum obtained when the sample was in place. The results are shown in Fig. 5 [Figure 5: see original paper], where “sample-in”denotes the effect spectrum measured with the sample, and “sample-out” represents the background spectrum measured without the sample. The effect-to-background ratio reached a peak value of 28 with an integrated effect-to-background ratio of 15 across the entire TOF spectrum.

III. MONTE CARLO SIMULATION

Shielding integral experiments validate nuclear data by comparing experimentally measured leakage neutron spectra with simulation results based on evaluated nuclear data libraries. Accurate simulations are critical to the success of these experiments because discrepancies between simulation and experimental results directly indicate areas for improvement in nuclear data evaluations. Among various Monte Carlo simulation programs, MCNP is widely recognized for its advantages in data acquisition, material description, and geometric modeling.

MCNP simulations for shielding integral experiments involve detailed descriptions of the experimental setup, including geometry, material composition, source neutron parameters, and detector response functions. The TARGET program [34] was employed to calculate the angular distribution of the source neutron energy spectrum, and the NEFF program [35] was used to determine detection efficiency curves. The pulse time distribution of source neutrons was derived by inverting the TOF spectrum measured using the monitor [36, 37]. These comprehensive inputs ensured that simulations closely replicated experimental conditions, thereby providing a reliable basis for comparison.

A. Geometric Model

Accurate geometric modeling is essential for reliable simulations. Ideally, a detailed model including all experimental structures would produce the most accurate results. However, such models significantly increase computation time. To balance accuracy and efficiency, it is necessary to remove structures that have minimal impact on simulation results. This was achieved through a series of simulations.

First, a detailed geometric model of the experimental hall and measurement system was created based on actual dimensions and materials. The leakage neutron spectrum of the polyethylene sample was calculated and used as the reference spectrum in this study. Subsequently, a weighting method was applied to obtain leakage neutron spectra from various sections of the experimental hall. These sections included the front, rear, left, and right walls (with the beam direction as reference) as well as the ceiling and floor. In these simulations, wall weights were set to zero. Six spectra were generated and compared with the reference spectrum. Finally, ratios of the studied spectra to the reference spectrum were calculated. The results are shown in Fig. 6 Figure 6: see original paper, enabling analysis of the influence of different wall sections on the leakage neutron spectrum.

Analysis of the ratios revealed that the ceiling, floor, front, and rear walls had minimal impact on the leakage neutron spectrum, with deviations from the reference spectrum of less than 0.1%. The left wall primarily influenced the low-energy region of the leakage neutron spectrum, whereas the right side wall primarily affected the high-energy region.

To further investigate the effects of the left and right side walls, we divided them into different regions along the central axis of the collimated beam. Using the weighting method again, results showed that only the region within a 75 cm radius from the central axis of the side walls had a significant effect on simulation results, as shown in Fig. 6(b) and (c).

In summary, except for regions of the left and right side walls within a radius of 0.75 m, the influence of other hall walls on the leakage neutron spectrum can be neglected. These walls were excluded from the calculation model. The model considers only the geometric structure along the neutron path to the detector, which includes the target, sample, shadow bar, pre-collimator, wall collimator, and parts of the left and right walls. A schematic of the simplified model is shown in Fig. 7 [Figure 7: see original paper]. The geometric model was constructed using experimentally measured dimensions and material properties of the target, collimation components, and shielding walls, ensuring that simulation results closely matched the experimental setup and minimizing discrepancies caused by geometric inaccuracies.

B. Source Neutron Parameters

Source neutron parameters, including energy-angle distribution and pulse time distribution, play crucial roles in simulations. Accurate descriptions of these parameters are vital to ensuring consistency between simulated and experimental leakage neutron spectra. Any deviations in their description can lead to significant discrepancies.

In experiments using a D-T neutron source, not all incident D+ ions react with tritium in the target. Unreacted D+ ions are deposited on the target, leading to subsequent D-D reactions. Over time, as tritium in the target is depleted and more D+ ions accumulate, the neutron source becomes a mixed field of D-T and D-D neutrons with dynamically changing yields. The energy-angle distributions of D-T and D-D neutrons were calculated using the TARGET program, which incorporates target geometry and material parameters. The ratios of D-T and D-D neutron yields were determined from measured alpha particle and proton ratios obtained from associated particle detectors. These distributions were combined to produce source neutron spectra that accurately reflected experimental conditions.

The pulse time distribution of source neutrons was influenced by beam dynamics, such as initial energy dispersion, high-voltage ripple during acceleration, and deviations caused by beam clustering. These factors introduced slight changes in the source neutron emission profile over time. The TOF spectrum of source neutrons, measured by the monitor, was combined with simulated response matrices to calculate the pulse time distribution through inverse convolution, as shown in Eq. (2).

$$\begin{pmatrix} N_1 \\ N_2 \\ N_3 \\ \vdots \\ N_i \end{pmatrix} = \begin{pmatrix} R_{11} & R_{12} & R_{13} & \cdots & R_{1j} \\ R_{21} & R_{22} & R_{23} & \cdots & R_{2j} \\ R_{31} & R_{32} & R_{33} & \cdots & R_{3j} \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ R_{i1} & R_{i2} & R_{i3} & \cdots & R_{ij} \end{pmatrix} \begin{pmatrix} \Phi_1 \\ \Phi_2 \\ \Phi_3 \\ \vdots \\ \Phi_j \end{pmatrix}$$

where N_i represents the measured TOF spectrum, R is the response matrix (with elements R_{ij}), and Φ_j is the pulse time distribution. This approach provided a precise representation of the source neutron emission profile for subsequent simulations.

C. Detector Efficiency Curve

The energy distribution of neutrons leaking from the sample spans a broad range and requires comprehensive understanding of detector efficiency across this wide energy region. Because of limited availability of monoenergetic neutron sources, experimentally measuring detection efficiency across the entire energy spectrum is impractical. Instead, the NEFF program was used to calculate efficiency curves based on detector geometric parameters, photoresponse curve, and energy threshold. These calculated efficiency curves were validated against available experimental data for similar systems, ensuring their reliability for TOF spectrum normalization.

D. Simulation of Leakage Neutron TOF Spectrum

To evaluate the accuracy of Zr isotope data in various nuclear data libraries, we replaced the natZr material cards in MCNP simulations. natZr consists of isotopes ^{90}Zr, ^{91}Zr, ^{92}Zr, ^{94}Zr, and ^{96}Zr, with proportions of 51.54%, 11.22%, 17.15%, 17.38%, and 2.8%, respectively. Evaluated nuclear data for these isotopes were sourced from the CENDL-3.2, ENDF/B-VIII.0, JEFF-3.3, and JENDL-5 libraries. The ENDF/B-VIII.0 library was used for all other materials, including Cu in the target structure, polyethylene, Fe, and Pb in the collimation system, ensuring that any differences in TOF spectra originated solely from natZr nuclear data.

After normalization using calculated detector efficiency curves, simulated TOF spectra were directly compared with experimental results, providing insights into the accuracy of nuclear data evaluations.

IV. RESULTS AND DISCUSSION

A. Verifying the System with a Standard Sample

To ensure reliability of the experimental measurement and data acquisition systems, we conducted a validation study by comparing simulated and experimental results of n-p scattering peaks using a polyethylene sample. Leakage neutron spectra from the polyethylene sample were experimentally measured at scattering angles of 47°, 61°, and 79°. The measured spectra were compared with simulated results obtained using four evaluated nuclear data libraries: CENDL-3.2, ENDF/B-VIII.0, JEFF-3.3, and JENDL-5. The results are shown in Fig. 8 [Figure 8: see original paper]. Close agreement between experimental measurements and simulated results confirmed the accuracy and reliability of the measurement system, data acquisition setup, and subsequent data processing procedures employed throughout the study.

B. Uncertainty Analysis

The experimentally measured TOF spectrum of leaked neutrons must be normalized before comparison with simulation results. The normalized TOF spectrum is given by Eq. (3):

$$TOF_{\text{final}} = \frac{TOF_{\text{out}}}{N_n \times S} = \frac{TOF_{\text{in}} \times \epsilon_{EN}}{(K_\alpha \times N_\alpha + K_p \times N_p) \times S}$$

where: - TOF_{out} denotes the neutron TOF spectrum count detected by the detector, which equals the actual neutron TOF spectrum count TOF_{in} multiplied by detector efficiency ϵ_{EN}. - N_n denotes the source neutron count, which equals detected alpha particle counts N_α and proton counts N_p multiplied by corresponding coefficients K_α and K_p, then summed. - S represents the detector area.

Therefore, the main sources of error include: - Statistical error of each time bin of the neutron TOF spectrum from experimental measurements ($< 3\%$). - Error of the associated particle monitoring system ($< 3\%$). - Statistical error of alpha particle counts and proton counts ($< 0.2\%$). - Detector efficiency error ($< 3\%$). - Stereoscopic angle error caused by experimental sample and detector placement ($< 1\%$).

C. Measurement Results of the Zr Samples

The leakage neutron TOF spectra of natZr samples with three thicknesses at six angles were measured in the experiment, resulting in 18 sets of data. Results for the three thicknesses are shown in Figs. 9–11 for comparison with leakage neutron spectra simulated by MCNP based on CENDL-3.2, ENDF/B-VIII.0, JEFF-3.3, and JENDL-5 libraries.

Figure 9 [Figure 9: see original paper]. (Color online) Comparison between calculated and measured leakage spectra of the 6 cm natZr sample.

By comparing calculated and experimental leakage neutron spectra, the following observations were made:

1. At approximately 156 ns, simulation results from the CENDL-3.2 library slightly underestimated experimental results at small angles (below 79°) but significantly overestimated them at large angles.
2. At approximately 164 ns, simulation results from CENDL-3.2 and JEFF-3.3 libraries were higher than experimental results across all angles, whereas simulation from ENDF/B-VIII.0 library was lower than experimental results at small angles (47°, 58°, and 61°).
3. Simulation results for CENDL-3.2 and JEFF-3.3 libraries were lower than experimental results for 300 ns–500 ns.

Figure 10 [Figure 10: see original paper]. (Color online) Comparison between calculated and measured leakage spectra of the 12 cm natZr sample.

Figure 11 [Figure 11: see original paper]. (Color online) Comparison between calculated and measured leakage spectra of the 18 cm natZr sample.

1. Change in C/E Values with TOF C/E values were obtained by dividing simulation results from the CENDL-3.2 library by experimental data. Variation in C/E with TOF is shown in Fig. 12 [Figure 12: see original paper]. As indicated, between 145 and 160 ns at small angles (47°, 58°, and 61°), simulation results were slightly lower than experimental values, whereas at larger angles they were clearly higher. Near 164 ns, simulation results exceeded experimental values; however, between 250 and 550 ns, simulation results were lower than experimental values.

Similarly, C/E values were obtained by dividing simulation results from the ENDF/B-VIII.0 library by experimental data. Variation in C/E with TOF is shown in Fig. 13 [Figure 13: see original paper]. As the figure shows, between 145 and 160 ns, simulation results were higher than experimental values; however, between 160 and 250 ns, most simulation results were lower than experimental values.

For JEFF-3.3, variation in C/E with TOF is shown in Fig. 14 [Figure 14: see original paper]. As shown, between 145 and 160 ns, simulation results were higher than experimental values, particularly at smaller angles ($\leq 79°$). Between 300 and 500 ns, simulation results were lower than experimental values, and this discrepancy was even more pronounced at larger angles.

For the JENDL-5 library, variation in C/E with TOF is shown in Fig. 15 [Figure 15: see original paper]. As shown, between 145 and 150 ns at smaller angles ($< 101°$), simulation results exceeded experimental values. Between 220 and 400 ns at larger angles ($> 119°$), simulation results were lower than experimental values.

2. Comparison of C/E Values in Different Energy Regions Discrepancies exist between simulations from different libraries and experimental results. These differences are caused by variations in reaction cross-sections related to secondary neutron production in each library. Analyzing these discrepancies can help identify potential limitations within libraries.

Using NDPlot [38] software developed by the China Nuclear Data Center, we obtained secondary neutron energy spectra and energy spectra of different reaction channels produced by 14.5 MeV neutrons incident on natZr samples from the CENDL-3.2 library. The results, shown in Fig. 16 [Figure 16: see original paper], were derived by inputting the proton number, mass number of the target nucleus, and energy of the incident particle.

According to contributions from different reaction channels to the total cross-section, the entire energy region can be divided into the following intervals:

- Elastic scattering interval (n, el): 13.4–15 MeV
- Discrete energy level inelastic scattering interval (n, inl)D: 10.6–13.4 MeV
- Continuous energy level inelastic scattering interval (n, inl)C: 4.0–10.6 MeV

- (n, 2n) reaction interval: 0.8-4.0 MeV

These energy intervals correspond to TOF intervals at different angles, as listed in Table 3 .

Table 3. TOF regions of each reaction channel at different angles.

Angle / °	(n, el) / ns	(n, inl)D / ns	(n, inl)C / ns	(n, 2n) / ns

To analyze cross-section data from different reaction channels, we calculated C/E values for measurements of samples with different thicknesses at different angles and compared them with simulation results from CENDL-3.2, ENDF/B-VIII.0, JEFF-3.3, and JENDL-5 libraries, as shown in Table 4 .

Table 4. C/E value of each reaction channel for four libraries.

Thickness-Angle	Reaction channel	CENDL-3.2	ENDF/B-VIII.0	JEFF-3.3	JENDL-5
6 cm-47°	(n, el)	0.923 ± 0.030	1.183 ± 0.039	1.152 ± 0.037	1.093 ± 0.036
6 cm-47°	(n, inl)D	1.106 ± 0.038	0.764 ± 0.026	1.128 ± 0.038	1.040 ± 0.035
6 cm-47°	(n, inl)C	0.902 ± 0.029	0.908 ± 0.029	0.998 ± 0.032	1.032 ± 0.033
6 cm-47°	(n, 2n)	0.917 ± 0.029	0.980 ± 0.031	0.940 ± 0.030	1.002 ± 0.032
...	...	...	...	...	...

3. Variation in C/E Values with Angles Variation in C/E values with angle for different reaction channels of various thicknesses is illustrated in Fig. 17 [Figure 17: see original paper], from which the following can be observed:

1. In the elastic scattering energy range, C/E values for ENDF/B-VIII.0, JEFF-3.3, and JENDL-5 libraries showed little variation with angle. In contrast, C/E values from the CENDL-3.2 library increased as angle increased. Although initially lower than those of the other three databases at smaller angles (47°, 58°, 61°, and 73°), C/E values from CENDL-3.2 library eventually surpassed those at larger angles.
2. In the discrete-level inelastic scattering energy range, C/E values from the ENDF/B-VIII.0 library were noticeably lower than those from the

other three libraries at all angles. Except for 122° and 133°, C/E values of CENDL-3.2 and JENDL-5 libraries remained close to each other and exceeded 1 across all angles.

3. In the continuous-level inelastic scattering energy range, C/E values of CENDL-3.2 and ENDF/B-VIII.0 libraries exhibited a slight increasing trend with angle, whereas those of JEFF-3.3 and JENDL-5 libraries exhibited a slight decreasing trend. At 101° and 107°, C/E values from all four libraries were most similar.
4. In the (n, 2n) energy range, C/E values of all four libraries exhibited little angular dependence. Compared with ENDF/B-VIII.0 and JENDL-5 libraries, C/E values from CENDL-3.2 and JEFF-3.3 libraries were relatively lower.

4. Analysis and Discussion To further analyze causes of discrepancies in C/E values observed across different databases, we employed the NDplot program to obtain secondary neutron energy spectra, double-differential cross-sections, and angular distributions for various reaction channels resulting from 14.5 MeV neutrons incident on natZr, as illustrated in Fig. 18 [Figure 18: see original paper].

1. Angular distribution curves of the (n, el) reaction were extracted from all four libraries and compared (Fig. 18(a)). At smaller angles, cross-section values from ENDF/B-VIII.0, JENDL-5, and JEFF-3.3 libraries were significantly higher than those from CENDL-3.2 library. However, at larger angles, CENDL-3.2 library exhibited higher cross-section values than the other three. This was likely the main reason why, in the elastic scattering energy range, simulations from all libraries except CENDL-3.2 overestimated experimental results at small angles, and all four libraries overestimated at larger angles.
2. Figure 18(b) shows angular distribution curves of the (n, inl)D reaction extracted from the four libraries. The (n, inl)D reaction cross-section was obtained by summing cross-sections of the 40 reaction channels (MT = 51 to 90). At all angles, cross-section values from CENDL-3.2 and JEFF-3.3 libraries were relatively high, whereas those from ENDF/B-VIII.0 library were the lowest. This likely explains why simulation results from CENDL-3.2 and JEFF-3.3 libraries in the discrete-level inelastic scattering energy range were higher than experimental values, whereas those from ENDF/B-VIII.0 library were lower.
3. Angular distribution curves of the (n, inl)C reaction extracted from the four libraries are shown in Fig. 18(c). Cross-section values of JEFF-3.3 library decreased noticeably with increasing angle. This was likely the main reason simulations from JEFF-3.3 library underestimated experimental results at large angles in the continuous-level inelastic scattering energy range.

4. Figure 18(d) shows (n, 2n) reaction energy spectra extracted from the four libraries. Cross-section values from CENDL-3.2 and JEFF-3.3 libraries were lower than those from ENDF/B-VIII.0 and JENDL-5 libraries. This is likely the main reason why simulation results from CENDL-3.2 and JEFF-3.3 libraries were lower than experimental results in the (n, 2n) energy range.

V. CONCLUSION

Using the CIAE shielding integral experiment measurement system, leakage neutron time-of-flight spectra were measured for 14.5 MeV neutrons generated by D–T fusion incident on natZr slabs with thicknesses of 6, 12, and 18 cm. Measurements were performed at angles of 47°, 58° (61°), 73° (79°), 107° (101°), 122° (119°), and 133°.

The MCNP program was employed to build a geometric model reflecting the experimental setup, considering angular distribution and pulse time distribution of source neutrons as well as detector efficiency. Leakage neutron spectra were calculated based on cross-section data for natZr isotopes in CENDL-3.2, ENDF/B-VIII.0, JEFF-3.3, and JENDL-5 libraries. Comparison between experimental and calculated results showed that simulations from different libraries exhibited varying degrees of deviation from measurements.

The secondary neutron energy spectrum produced by 14.5 MeV incident neutrons on natZr samples was obtained using the NDplot program, and the whole energy spectrum interval was divided into the elastic scattering interval (n, el), discrete energy level inelastic scattering interval (n, inl)D, continuous energy level inelastic scattering interval (n, inl)C, and (n, 2n) interval according to contributions from different reaction channels to the total cross-section.

The C/E value was obtained by dividing calculated values by experimental values. Analyzing C/E values for different energy regions, we obtained the following conclusions: In the (n, el) energy region, the C/E value for CENDL-3.2 library increased with angle. At small angles, simulated results from CENDL-3.2 were slightly lower than experimental values, whereas at large angles they were significantly higher. For the other three libraries, C/E values did not vary significantly with angle, and simulated results were consistently higher than experimental results. From the secondary neutron angular distribution for this reaction channel, cross-section values from CENDL-3.2 library were lower than those from the other three libraries at small angles and higher at large angles. In the (n, inl)D energy region, simulated results from CENDL-3.2 and JEFF-3.3 libraries were consistently higher than experimental results at all angles, whereas ENDF/B-VIII.0 library exhibited significantly lower simulation results. Cross-section values for ENDF/B-VIII.0 were notably lower than those of the other three libraries across all angles. In the (n, inl)C energy region, JEFF-3.3 library simulation results were lower than experimental values at large angles. The secondary neutron angular distribution for this reaction channel showed that cross-section values from JEFF-3.3 decreased with angle and were much lower than those from other databases at large angles. In the (n, 2n) energy region, simulated results from both CENDL-3.2 and JEFF-3.3 libraries were lower than experimental results. From the secondary neutron energy spectrum of this reaction channel, cross-section values for these two libraries were lower than those for the other two libraries.

These differences indicate that discrepancies still exist in evaluated nuclear data for Zr across different libraries, particularly in the (n, el) and (n, inl)D reaction energy regions, where differences are most pronounced. Further integral experiments are required to provide data support for improving evaluated nuclear data of Zr.

In summary, results of this study highlight the necessity of enhancing accuracy and consistency of zirconium-evaluated nuclear data and provide valuable insights for improvement.

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AUTHOR CONTRIBUTIONS

All authors contributed to study conception and design. Material preparation, data collection, and analysis were performed by Shiyu Zhang, Yanyan Ding, and Qi Zhao. The first draft of the manuscript was written by Xinyi Pan, and all authors commented on previous versions. All authors read and approved the final manuscript.

DATA AVAILABILITY

The data that openly support the findings of this study are available at https://www.doi.org/10.57760/sciencedb.j00186.00987 and https://cstr.cn/31253.11.sciencedb.j00186.00987.

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CONFLICT OF INTEREST

The authors declare that they have no competing interests.

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