Neutron Response and Energy Spectrum Measurement within 0.1-300 MeV Based on a CLYC Detector

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Abstract

Cs2LiYCl6: Ce (CLYC) scintillator is sensitive to neutrons within a wide-energy range and has the potential to achieve wide-range neutron energy spectrum measurements. The direct neutron detection performance of a CLYC detector has been realized by utilizing the Back-n white neutron source at China Spallation Neutron Source (CSNS), where a 20 cm lead brick was designed to effectively shield the accompanying gamma flash from the neutron source. The detected neutron energy spectrum after passing through the lead brick is primarily distributed within the 0.1–300 MeV and exhibits identical four-peak structures, which are consistent with Geant4 simulation results. The peak energies of the 1st, 3rd, and 4th peaks match well, with the relative deviation being less than 6%. However, a large energy deviation has been found for the 2nd peak whose energy falls within the resonance energy region of lead, which indicates that the resonance reaction of lead might not be precisely simulated in Geant4. Due to the dominance of elastic scattering between neutrons and the main nuclides (6Li, 35Cl, 37Cl, 89Y, 133Cs, 140Ce) in the CLYC scintillator in the low-energy region, suppression has been found for reactions that are used for neutron detection reactions such as (n, p), (n, d), (n, t) and (n, α). This leads to a significant difference in overall intensity between the low-energy range within 0.1–3 MeV (1st and 2nd peaks) and the high-energy range within 5–64 MeV (3rd and 4th peaks). A large number of gamma signals were measured in the experiment, which were found to be prompt gamma rays generated from neutron reactions with CLYC, but not the associated gamma rays from the neutron source. This phenomenon is more pronounced for the high-energy region, these prompt gamma rays can be combined into the neutron pulses and render the neutron pulses to contain fast-decay components that originally only existed in gamma pulses, and further decrease the neutron gamma discrimination performance of CLYC. By establishing a wide-energy neutron response matrix entirely based on experimental measurements and utilizing the GRAVEL unfolding method, the white neutron energy spectrum in the range of 6–200 MeV was successfully unfolded. These results provide a critical reference for the accurate measurement and discrimination of neutron-gamma signals in radiation fields using CLYC detectors and indicate the feasibility of using CLYC detectors for wide-energy neutron spectrum measurements, highlighting its potential for applications in high-energy neutron experiments, space neutron detection, and other related fields.

Full Text

Preamble

Neutron Response and Energy Spectrum Measurement within 0.1–300 MeV based on a CLYC Detector

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The Cs_2LiYCl_6:Ce (CLYC) scintillator exhibits sensitivity to neutrons across a wide energy range and demonstrates potential for broad-spectrum neutron energy measurements. The direct neutron detection performance of a CLYC detector has been evaluated using the Back-n white neutron source at the China Spallation Neutron Source (CSNS), where a 20 cm lead brick was designed to effectively shield the accompanying gamma flash from the neutron source. The detected neutron energy spectrum after passing through the lead brick is primarily distributed within 0.1–300 MeV and exhibits four distinct peak structures that are consistent with Geant4 simulation results. The peak energies of the 1st, 3rd, and 4th peaks match well, with relative deviations less than 6%. However, a large energy deviation was found for the 2nd peak, which falls within the resonance energy region of lead, indicating that the resonance reaction of lead might not be precisely simulated in Geant4. Due to the dominance of elastic scattering between neutrons and the main nuclides (^{6}Li, ^{35}Cl, ^{37}Cl, ^{89}Y, ^{133}Cs, ^{140}Ce) in the CLYC scintillator in the low-energy region, reactions used for neutron detection such as (n, p), (n, d), (n, t) and (n, α) are suppressed. This leads to a significant difference in overall intensity between the low-energy range within 0.1–3 MeV (1st and 2nd peaks) and the high-energy range within

5–64 MeV (3rd and 4th peaks). A large number of gamma signals were measured in the experiment, which were found to be prompt gamma rays generated from neutron reactions with CLYC, not associated gamma rays from the neutron source. This phenomenon is more pronounced in the high-energy region, where these prompt gamma rays can combine with neutron pulses and render them to contain fast-decay components that originally existed only in gamma pulses, further decreasing the neutron-gamma discrimination performance of CLYC. By establishing a wide-energy neutron response matrix entirely based on experimental measurements and utilizing the GRAVEL unfolding method, the white neutron energy spectrum in the range of 6–200 MeV was successfully unfolded. These results provide a critical reference for accurate measurement and discrimination of neutron-gamma signals in radiation fields using CLYC detectors and demonstrate the feasibility of using CLYC detectors for wide-energy neutron spectrum measurements, highlighting their potential for applications in high-energy neutron experiments, space neutron detection, and other related fields.

Keywords: CLYC detector, Wide-energy neutron, Neutron response, Neutron energy spectrum measurement

Introduction

Since the first discovery of the excellent performance of Cs_2LiYCl_6:Ce (CLYC) scintillation by C. M. Combes's group in 1999, extensive research has been conducted on this novel scintillator over the past two decades. Researchers have found that CLYC detectors can detect both thermal and fast neutrons through interactions with ^{6}Li and ^{35}Cl, respectively, and CLYC is also capable of detecting high-energy neutrons via heavy nuclides such as ^{133}Cs and ^{89}Y.

In addition, the CLYC scintillator exhibits better energy resolution (about 5% @ 662 keV) than the widely used NaI(Tl) crystal. Moreover, owing to its Core-to-Valence Luminescence (CVL) mechanism, CLYC enables efficient simultaneous detection of neutrons and gamma rays. These superior properties endow CLYC scintillators with great potential for a wide range of applications in multimodal neutron-gamma detection. The wide-energy neutron response capability of CLYC provides significant advantages in neutron energy spectrum measurements.

However, current studies on the neutron response of CLYC are mostly focused on the low-energy neutron region, while studies on high-energy neutrons, especially those above 10 MeV, remain insufficient. This limitation constrains the application of CLYC in high-energy neutron experiments, space neutron measurements, and other related fields. Moreover, in neutron energy spectrum measurements, due to the limitations of monoenergetic neutron sources, the neutron response function of the detector is typically obtained using Monte Carlo simulation methods (such as FLUKA or GEANT4), and multiple response functions are then combined to construct a response matrix. However, for CLYC detectors, it is difficult to accurately obtain the neutron response function using simulation methods, which can significantly affect the final neutron energy spectrum. In the actual reaction process, quenching effects occur which cause the number of scintillation photons to be inconsistent with the deposited energy of the neutron reaction products. Additionally, as neutron energy increases, more nuclear reaction channels become available, and different reaction channels at different neutron energies correspond to different quenching factors, making the actual scintillation process even more complex. Therefore, in simulations, it is necessary to incorporate experimentally determined quenching factors corresponding to these complex nuclear reactions to realistically model the quenching process and obtain a more accurate neutron response function. Nevertheless, experimental studies on the quenching factors of different nuclear reactions in CLYC are very limited and mostly restricted to low-energy neutrons. Furthermore, the interactions between high-energy neutrons and CLYC are extremely complex, making it extremely difficult to realistically model the quenching processes in simulations. Consequently, it is necessary to experimentally establish the actual response matrix of CLYC to enable spectral measurements of neutrons over a wide-energy range and to further study the characterization of CLYC response to wide-energy, particularly high-energy neutrons.

This study established an experimental design to obtain the actual response matrix of CLYC by using the Back-n white neutron source at CSNS in conjunction with the Geant4 simulation toolkit. Both simulations and experimental measurements were employed to study the influence of lead bricks in the experimental design on the Back-n white neutron energy spectrum. In addition, the neutron response of the CLYC detector in the energy range from 1 eV to 400 MeV was studied, and the results were interpreted based on Geant4 simulations and nuclear reaction cross sections. Finally, an experimental wide-energy neutron response matrix was established, and the GRAVEL method was applied to achieve the unfolding of the white neutron energy spectrum in the 6–200 MeV energy range.

2. Measurement Setup

The CLYC detector consists of a $\Phi 1$ in $\times$ 1 in CLYC scintillator (using ^{6}Li enrichment of 95%) coupled with a Hamamatsu R6231-100 photomultiplier tube (PMT). The pulse signals were acquired using a high-speed digitizer with a sampling rate of 1 GHz and a vertical resolution of 12 bits. To improve the accuracy of neutron energy measurement by the time-of-flight (TOF) method, the CLYC detector was placed at the second end-station hall (ES#2), which was further away from the target station (the flight path was about 77.58 m). To acquire wide-energy neutrons from keV to hundreds of MeV via the TOF method, the sampling window was set to 25,000 ns, and the digitizer was configured in external trigger mode (the trigger signal was provided by CSNS).

In this work, the signal obtained by each sampling window is defined as an event. The Back-n white neutrons are produced via the spallation reaction of a 1.6 GeV proton beam impinging on a tungsten target at a repetition rate of 25 Hz, and the resulting neutron beam is separated from the proton beam by a 15° bending magnet. This study was conducted at CSNS in the double-bunch mode, in which each proton beam pulse contains two bunches with a time interval of 410 ns, and the full width at half maximum (FWHM) of each bunch is approximately 42 ns. Furthermore, to accurately obtain the pulse integration spectrum of the CLYC detector in response to neutrons, it is necessary to minimize the occurrence of pile-up pulses. Therefore, the smallest diameter combination of the shutter (Φ3 mm), collimator1 (Coll#1, Φ15 mm) and collimator2 (Coll#2, Φ40 mm) was selected.

It should be noted that there is large production of gamma rays when the proton beam bombards the spallation target. They can be classified into two categories: the prompt gamma rays (γ-flash), which have the same time structure as the proton beam; and the delayed gamma rays, which don't have evident time structure and are decayed gamma rays from nuclei produced by neutron-induced reactions and spallation reactions. Consequently, lead bricks with a thickness of 20 cm were placed in the first end-station hall (ES#1) to shield these gamma rays (see Section 2.2 for details). The experimental setup is illustrated in Fig. 1 [Figure 1: see original paper]. The data were analyzed, fitted, and mapped using the ROOT package developed by CERN.

Due to the inherent range (-20 mV, 480 mV) of the digitizer, the operating voltage of the PMT was optimized to 900 V to obtain a wider energy range of neutron pulse signals. At this voltage, the energy resolution of the CLYC detector for 0.662 MeV gamma rays from ^{137}Cs was studied under different integration time lengths ranging from 1000 ns to 5000 ns, with a step size of 1000 ns. The optimal integration time was determined to be 2000 ns, yielding an energy resolution of 7.56% at 0.662 MeV. This resolution is worse than the best value of 5% for this system when the PMT's operating voltage is much higher. Moreover, the energy calibration was done using four reference points: the 0.662 MeV gamma ray peak from a ^{137}Cs source, the 0.778 MeV and 1.408 MeV gamma ray peaks from a ^{152}Eu source, and the approximately 3.2 MeV gamma equivalent energy (GEE) of the slow neutron peak from a ^{241}Am-^{9}Be source moderated by paraffin. As shown in Fig. 2 [Figure 2: see original paper], the CLYC detector exhibited excellent energy linearity across this energy range.

2.1 Start Time of Neutron TOF

Before obtaining the response data of the CLYC detector to neutrons over a wide-energy range, it is necessary to determine the start time (T_0) of the neutron TOF. Although the external trigger signal provided by CSNS can be used as T_0, there may be an unknown time delay between this trigger signal and the exact moment when the protons hit the target, which might originate from electronics and cable connection delays. Therefore, in this study, the γ-flash pulse signal was used to obtain an accurate T_0. The relevant information of the γ-flash pulse signal was obtained using a peak-seeking algorithm developed by our group in a previous study. Under conditions without lead bricks, a typical example of the γ-flash pulse signal in the double-bunch mode is shown in Fig. 3 [Figure 3: see original paper], and the start time of the two γ-flash pulses is illustrated. As seen in the figure, the second γ-flash pulse signal exhibits saturation, which can be attributed to two main reasons. First, the intensity of the γ-flash is very high, and the pulse has a long tail. The γ-flash pulse signal from the second bunch is superimposed on the falling edge of the first γ-flash pulse signal, resulting in signal saturation. Second, in the double-bunch mode, the interval between the second and first proton bunches is 410 ns. During the detection period of the second γ-flash, some high-energy neutrons generated by the first proton bunch have arrived at the CLYC detector. The resulting neutron-induced pulse signals may overlap with the γ-flash signals from the second proton bunch, further contributing to signal saturation. Approximately 1.6×10^4 γ-flash events are detected in the two-bunch mode, and the distribution of the start time for the 1st and 2nd γ-flash pulses are shown in Fig. 4 [Figure 4: see original paper]. Gaussian fitting acquired the average start time of the two γ-flash signals as 3531 ns (t_1) and 3941 ns (t_2), respectively. The interval between these two times is 410 ns, which is consistent with the double-bunch time interval provided by CSNS. This result also demonstrates the good accuracy of the peak-seeking algorithm used. According to Eq. (1), the T_0 of the neutron TOF was calculated to be 3273 ns, where L is the flight path of 77.58 m, and c is the speed of light.

3. Shielding Lead Brick Optimization

When acquiring the response data of the CLYC detector to neutrons over a wide-energy range, lead bricks were placed at ES#1 to prevent excessive gamma rays and significant pulse pile-up, thereby minimizing their impact on neutron energy spectrum measurements and neutron response studies. To determine the optimal lead brick thickness, Geant4 simulations were conducted for four configurations: 6 cm, 10 cm, 15 cm, and 20 cm. A planar gamma ray source, whose beam spot diameter is Φ15 mm, was modeled by sampling the energy spectrum shown in Fig. 5 [Figure 5: see original paper], which is provided by CSNS. A total of 2×10^7 particles were sampled, and the gamma particles that passed through the lead brick and emitted along the beam direction (0°) were recorded. These gammas included both primary gammas that passed through the lead brick and secondary gamma rays generated via interactions within the lead brick. These results are shown in Table 1 . The reason that only gamma rays along the 0° direction were recorded is that the distance between the lead brick at ES#1 and the CLYC detector at ES#2 is approximately 21 m, and the solid angle subtended by the CLYC detector is negligible when compared to this distance. Additionally, the thick wall between ES#1 and ES#2 shields gamma rays emitted at other angles, so only gamma rays emitted along the beam direction (0°) can be detected by the CLYC at ES#2. As shown in Table 1, with a 20 cm thickness of lead bricks, gamma rays are almost completely shielded, reducing transmitted particles to 0.00027% of the source particles.

In the experiment, due to limited beam time acquired from the Back-n white neutron source at CSNS, tests under lead brick thicknesses of 6 cm, 15 cm, and 20 cm were studied for time efficiency and operational feasibility. Under the 6 cm lead brick configuration, extensive pulse pile-up occurred, which caused baseline distortions for signals that followed the first pulse in each event. This prevented accurate calculation of the pulse integral spectrum and hindered neutron-gamma pulse discrimination. Representative pulse signals for 6 cm and 15 cm thick lead bricks are illustrated in Fig. 6 [Figure 6: see original paper]. When the lead brick is thicker, the pile-up becomes less severe, and the number of pulses within an event can be discriminated after using the peak-seeking algorithm. The number of pulse distributions for 15 cm and 20 cm lead bricks are shown in Fig. 7 [Figure 7: see original paper] (a) and (b), respectively. The number of valid events is similar for both thicknesses, with 11,634 for 15 cm and 11,614 for 20 cm. However, the 20 cm configuration exhibits significantly higher single-pulse event counts compared to 15 cm. The proportions of single-pulse events are 41.02% and 72.80% for the 15 cm and 20 cm configurations, respectively. For multi-pulse events, subsequent pulses may overlap with the falling edges of preceding pulses, leading to baseline shifts that make it difficult to accurately determine the integral values of the pulses. In contrast, single-pulse events are more reliable for neutron energy spectrum measurement and response studies. Thus, employing a 20 cm thickness of the lead brick to attenuate gamma rays in the Back-n beamline allowed the accumulation of more single-pulse event data within limited beam time and facilitated subsequent research and analysis.

4. Results and Analysis

Experimental data spanning a total measurement duration of 16.5 hours were analyzed and neutron-gamma pulse discrimination was implemented using the stable and dependable charge comparison method, with optimized long and short integration windows of 400 ns and 70 ns, respectively. The variable R_{psd} is defined as the ratio of Q_{short} (integration of short window) to Q_{long} (integration of long window), as shown in Eq. (2). The two-dimensional scatter distribution of R_{psd} versus GEE values is shown in Fig. 8 [Figure 8: see original paper]. The figure demonstrates a distinct separation between neutron and gamma distributions, with gamma localized above the blue line and neutrons below. Within the neutron cluster, four prominent band structures and one relatively distinct band are observed. These measurement results align with those reported by R. S. Woolf et al., using a CLYC detector (enriched with ^{7}Li) under 6–60 MeV neutron irradiation. As neutron energy increases, multiple nuclear reaction channels such as (n, p), (n, d), (n, t), and (n, α) are opened for nuclides such as ^{35}Cl, ^{37}Cl, ^{89}Y, ^{133}Cs, ^{140}Ce and other nuclides in CLYC. At the same time, since neutrons of different energies induce different nuclear reaction channels, the reaction energy also varies accordingly. In addition, due to differences in the mass and charge of these emitted particles, the quenching effects are different for each channel, and the proportion of the slow-decay component is different for each channel which would influence the pulse shape characteristic. The combined and complex influence of these factors gives rise to these band regions. In Fig. 8, these bands correspond sequentially (top to bottom) to contributions from escaping protons, protons, deuterons, tritons, and α particles.

Due to these measurements being done in the double-bunch mode, the low-energy neutrons from the first bunch will be mixed with the high-energy neutrons from the second bunch. To further study the relationship between the GEE and neutron TOF, the double-bunch unfolding algorithm developed by CSNS was employed to obtain equivalent measurement results corresponding to the single-bunch mode. The relationship between neutron GEE and TOF is shown in Fig. 9 [Figure 9: see original paper].

As seen in Fig. 9 (a), the original spectrum contains two overlapped bunch distributions (bunch 1 and bunch 2). Fig. 9 (b) shows the equivalent single-bunch distribution after applying the double-bunch unfolding algorithm. Because the unfolding performance in each energy interval depends on the data statistics, in the region where GEE exceeds 65.4 MeV, the original spectrum contains relatively few counts per energy interval, resulting in significant fluctuations in the unfolding results in this region. However, since this energy range accounts for only 0.95% of the total data, these fluctuations have a negligible effect on the overall distribution and subsequent analysis. From Fig. 9 (c), it can be observed that, in the band region corresponding to the ^{35}Cl(n, p)^{35}S reaction, there are two distinct overlapping bands (bunch 1 and bunch 2) before unfolding. After the double-bunch unfolding, the influence of the second bunch is effectively eliminated, as shown in Fig. 9 (d). Furthermore, three clearly separated regions are identified in Fig. 9 (d), corresponding to the ^{6}Li(n, t)α reaction ( ), the ^{35}Cl(n, p)^{35}S reaction ( ), and complicated reactions ( ) induced by higher energy neutrons with the main nuclides (^{6}Li, ^{35}Cl, ^{37}Cl, ^{89}Y, ^{133}Cs, and ^{140}Ce) in the CLYC scintillator. Notably, as observed in Fig. 9 (d), the band structures corresponding to the ^{6}Li(n, t)α and ^{35}Cl(n, p)^{35}S reactions progressively merge into the complex nuclear reaction regions of higher energy neutrons as the TOF decreases. This observation demonstrates the viability of ^{6}Li and ^{35}Cl for detecting high-energy neutrons.

4.1 Neutron Energy Spectrum

This study employed a 20 cm thick lead brick for gamma ray shielding, but this thickness inevitably influenced the white neutron energy spectrum of Back-n, thereby altering the incident neutron energy spectrum structure entering the CLYC detector. To address this issue, Geant4 simulations were conducted by utilizing the Back-n white neutron energy spectrum provided by CSNS, sampling 5×10^7 source neutron particles. It should be noted that CSNS does not provide energy spectrum data for the shutter, Coll#1 and Coll#2 under the Φ3 mm, Φ15 mm, and Φ40 mm combination used in this study. Instead, the neutron energy spectrum under the Φ12 mm, Φ15 mm, and Φ40 mm combination, which is the closest configuration to the one used in this work, was adopted as provided by CSNS. Analysis of the energy spectra across various configurations supplied by CSNS reveals no significant structural differences in the neutron energy spectra within the 0.1–300 MeV range. The resultant neutron energy spectrum distribution along the 0° direction (same direction as the incident neutron source) after lead (Pb) shielding was statistically analyzed, as illustrated in Fig. 10 [Figure 10: see original paper]. The total neutron reaction cross section of Pb shown in panel (b) was obtained from the IAEA nuclear cross section database ENDF/B-VIII.0. The neutron energy spectrum entering the CLYC detector exhibits four prominent peaks (, , ,) after 20 cm Pb attenuation. The first sharp and narrow peak ( ) is attributed to the abrupt decrease in the total cross section of Pb at approximately 0.49 MeV. The second peak ( ), situated within the resonance region of Pb's neutron total cross section, exhibits less distinct peak characteristics. Peaks and are caused by the spectral valleys in Pb's neutron total cross section corresponding to neutron energy ranges of approximately 5–16 MeV and 27–64 MeV, respectively.

Utilizing the TOF method, as shown in Eq. (3), the energy spectrum of the neutrons of Fig. 8 was obtained, and after double-bunch unfolding, the resulting spectrum is shown in Fig. 11 [Figure 11: see original paper] (a). Meanwhile, in Geant4 simulations, 2×10^7 source particles were sampled according to the neutron energy spectrum distribution shown in Fig. 10 (b). For neutron-induced pulse signals in the CLYC detector, only the (n, p), (n, d), (n, t), and (n, α) reactions were considered. This is because heavier ions (such as ^{6}Li) exhibit stronger quenching effects in scintillators, leading to greater non-radiative energy losses and significantly reduced scintillation light yield, making it difficult to generate detectable pulse signals. By statistically analyzing source neutron events that induced (n, p), (n, d), (n, t), and (n, α) reactions with deposited energy greater than zero, the neutron energy distribution is shown in Fig. 11 (b).

As shown in Fig. 11, both experimental and simulated results exhibit four distinct peaks. The energy ranges of these peaks align with those in Fig. 10 (b), which are attributed to the 20 cm lead brick. Gaussian fitting was applied to determine the centroid energies of these peaks, and the absolute relative deviations between experimental and simulated peak positions are summarized in Table 2 . The distribution range of peak is relatively narrow and distinct, with no deviation between the experimental and simulated results. For peak , since the incident neutron energy spectrum in this energy range corresponds to the resonance cross section region of Pb, the peak position is not pronounced and the fitting results show a large deviation. The relative deviations between the experiment and simulation for peaks and are small. Overall, the peak positions in the experiment and simulation agree well, demonstrating the accuracy and validity of both the experimental measurements and simulation.

As shown in Fig. 10 (b), the overall intensity of incident neutrons in the energy ranges corresponding to Peaks and are the highest. However, both the experimental measurements and the simulated neutron response results indicate that the intensities of these peaks are the lowest, as shown in Fig. 11. The reaction channels between neutrons and various isotopes inside CLYC were simulated, including ^{6}Li, ^{35}Cl, ^{37}Cl, ^{89}Y, ^{133}Cs, and ^{140}Ce. It is found that the elastic scattering reactions occur most frequently, while (n, p), (n, d), (n, t), and (n, α) reactions are relatively rare. The contribution to energy spectra corresponding to these reactions were simulated and the results are shown in Fig. 12 [Figure 12: see original paper]. The cross sections for (n, p), (n, d), (n, t), (n, α) and elastic scattering reactions between neutrons (0.1–20 MeV) and CLYC isotopes of ^{6}Li, ^{35}Cl, ^{37}Cl, ^{89}Y, ^{133}Cs, and ^{140}Ce are presented in Fig. 13 [Figure 13: see original paper] for comparison. The cross section data are primarily sourced from the IAEA nuclear data library ENDF/B-VIII.0, except for the ^{6}Li(n, d) reaction cross section, which is only available in the IAEA TENDL-2015.s60 database. Figure 12 reveals that the elastic scattering reactions are the most significant contribution to the four distinct peaks induced by the 20 cm lead brick. The intensities of Peaks and in elastic scattering (on the order of 10^4) are significantly higher than those in (n, p), (n, d), (n, t), and (n, α) reactions. From the perspective of reaction cross sections, the elastic scattering cross section in the energy ranges of Peaks and are far higher than that of (n, p), (n, d), (n, t), and (n, α) reactions. In the 0.1–2 MeV range, only the ^{6}Li(n, t) reaction contributes notably, yet its cross section remains lower than that of elastic scattering reactions. Above 2 MeV, the cross sections for ^{35}Cl(n, p) and ^{35}Cl(n, α) reactions gradually increase, while (n, p), (n, d), (n, t), and (n, α) reaction channels for ^{6}Li, ^{35}Cl, ^{37}Cl, ^{89}Y, ^{133}Cs, and ^{140}Ce progressively open. Therefore, the dominance of elastic scattering cross sections at lower neutron energies results in significantly reduced intensities of Peaks and of the (n, p), (n, d), (n, t), and (n, α) reactions when compared to that in the high-energy range (Peaks and). This observation further validates the effectiveness of CLYC detectors for high-energy neutron measurements.

As shown in Fig. 11, for high-energy neutrons above 20 MeV, the relative intensity of the measured energy spectrum is higher than that from the Geant4 simulation. The possible main reasons for this discrepancy are as follows: (1) The cross sections used in the Geant4 physical models for the high-energy neutron region may not be accurate; (2) The quenching effects of the CLYC scintillator are not precisely simulated. As the neutron energy increases, the number of nuclear reaction channels with various nuclides increases, resulting in a greater variety and number of emitted particles as well as differences in reaction energies. Consequently, the quenching factors become more complex at higher energies. At present, no studies provide accurate quenching factor values for these complex reactions in CLYC scintillators. Therefore, the simulated energy spectrum does not account for quenching effects from (n, p), (n, d), (n, t), and (n, α) reactions of neutrons with ^{6}Li, ^{35}Cl, ^{37}Cl, ^{89}Y, ^{133}Cs and ^{140}Ce, which may lead to differences in the relative intensity distributions between experiment and simulation; (3) To achieve accurate GEE calibration and minimize the impact on neutron energy spectrum measurement and response research, only single-pulse events were selected in the experiment when calculating the pulse integral spectrum. This event selection criterion will exclude multi-pulse events; under this condition, events with higher reaction probability might be selected more frequently and this will contribute to discrepancies between the measured and simulated energy spectra.

4.2 Gamma Response Analysis

As shown by the results in Section 2.2, the 20 cm lead brick almost completely shields all gamma rays. However, a large number of gamma pulse signals were observed in the actual measurements. Furthermore, Geant4 simulation results indicate that the secondary gamma particles produced by the lead brick do not enter the CLYC detector. To identify the origin of these measured gamma rays, detailed Geant4 simulations were carried out. The results reveal that some neutrons entering the CLYC undergo radiative capture reactions with ^{6}Li, ^{35}Cl, ^{37}Cl, ^{89}Y, ^{133}Cs, and ^{140}Ce, resulting in the emission of gamma rays. Another fraction of neutrons undergo inelastic scattering with these nuclides, exciting them, and these nuclides would emit gamma rays during their de-excitation. Inelastic scattering accounts for as much as 94.66% of these reactions. The gamma deposition spectra from these reactions in simulation and the experimentally measured gamma deposition spectra are shown in Fig. 14 [Figure 14: see original paper]. Due to the dynamic range setting of the experiment, many low-amplitude gamma pulse signals were not detected, so the experimental gamma deposition spectrum was almost entirely above 0.4 MeV. Therefore, the simulated spectrum was also truncated at a 0.4 MeV threshold. As shown in Fig. 14, the experimental and simulated gamma deposition spectra exhibit the same trend. In addition, Fig. 15 [Figure 15: see original paper] presents the measured TOF spectra of gamma and neutron pulse signals, together with the corresponding simulated spectra. The TOF spectra of gamma pulse signals in both the experimental and simulated results show a very similar structure to that of the neutron TOF spectra. Therefore, it can be concluded that these gamma pulse signals were produced by incident neutrons undergoing radiative capture and inelastic scattering with the main nuclides in CLYC, with inelastic scattering playing the dominant role. This result provides important experimental evidence for the accurate measurement of neutrons and gamma rays in complex radiation fields using CLYC detectors.

To evaluate the pulse shape of high-energy neutrons, typical pulse shape diagrams are shown in Fig. 16 [Figure 16: see original paper]. The neutron pulse signals were selected around 5 MeV, 10 MeV, 20 MeV, 30 MeV, and 40 MeV based on GEE. Each pulse signal was normalized, aligned at its starting point, accumulated and averaged, and then normalized to its maximum value. From the figure, it can be observed that as the GEE increases, the neutron pulse signals exhibit a fast-decay component similar to that of the gamma pulse signal, and the decay trend gradually approaches that of the gamma pulse signal. Combining this with Fig. 9 (b), it can be concluded that the pulse signals for GEE at energies higher than 10 MeV are predominantly caused by high-energy neutrons (>10 MeV). When high-energy neutrons undergo nuclear reactions in CLYC, they produce particles such as p, d, t and α, and may also cause residual nuclear excitation. The subsequent de-excitation of the nuclei releases gamma particles, which interact with CLYC and deposit energy, potentially resulting in gamma pulse signals. Then for a high energy neutron, the secondary gamma ray might be generated along with the secondary ions (p, d, t, α), which will render the neutron pulse signal to include a fast-decay component, and this possibility will be larger when neutron energy is higher. Consequently, as neutron energy increases, the difference between neutron and gamma pulses becomes smaller, and the discrimination becomes challenging when relying solely on the R_{psd} from the charge comparison method or other pulse shape discrimination methods.

4.3 White Neutron Energy Spectrum Unfolding

The response matrix (R) of the CLYC, which is used to calculate the detector's response (N) for neutron flux (ϕ), can be expressed in the discrete form of the first-kind Fredholm integral equation, as shown in Eq. (4):

$$N_i = \sum_{j=1}^{J} R_{ij}\phi_j$$

In Eq. (4), N_i represents the detector's count rate in the i-th bin of the measured spectrum; ϕ*j represents the flux in the j-th bin,* R{ij} is the element used to calculate the count rate of the i-th bin that contributed from the j-th incident neutron energy; and J is the total number of bins for the energy spectrum.

Once R is known, it can be used to unfold the unknown neutron energy spectrum (ϕ), when the detector's response (N) is measured. Because there are statistical characteristics for the detector's response N and the response matrix R, and a continuous response distribution can be found for the neutron with a fixed single energy, the neutron response matrix is usually ill-conditioned. Therefore, ϕ cannot be obtained by directly inverting the matrix. Instead, a spectrum unfolding method is required to solve the energy spectrum.

In this study, the R was derived from Fig. 9 (b) using Eq. (3) and is shown in Fig. 17 [Figure 17: see original paper]. Considering that the unfolding performance was affected by statistical counts, the R was constructed using responses within GEE of 0.4–65.4 MeV (step size 0.2 MeV) and neutron energy of 6–200 MeV (step size constrained by the CSNS double-bunch unfolding algorithm, corresponding to a TOF interval of approximately 29.4 ns). If the data acquisition time can be longer, the energy range of R can be further extended.

The GRAVEL method, which is a classical method for solving over-determined systems of linear equations, was considered to unfold the white neutron energy spectrum in this study. The initial energy spectrum for iterative unfolding was obtained by multiplying N with the transpose of the R. The iteration expression is as follows:

$$\phi_j^{(k+1)} = \phi_j^{(k)} \exp\left(\frac{\sum_{i=1}^{I} W_{ij}^{(k)} \ln\left(\frac{N_i}{Q_i^{(k)}}\right)}{\sum_{i=1}^{I} W_{ij}^{(k)}}\right)$$

where k indicates the number of iterations, I is the total number of bins of N, W_{ij}^{(k)} is a weight factor defined by:

$$W_{ij}^{(k)} = \frac{N_i R_{ij} \phi_j^{(k)}}{Q_i^{(k)}}$$

and Q_i^{(k)} is the reconstructed position distribution obtained by multiplying the response R_{ij} and the iterative solution ϕ_j^{(k)}, expressed as:

$$Q_i^{(k)} = \sum_{j=1}^{J} R_{ij} \phi_j^{(k)}$$

In addition, the GRAVEL method has issues such as incomplete convergence of the solution and a tendency for the solution to diverge with an increasing number of iterations. Therefore, it is necessary to introduce a criterion for stopping the iteration process. In this study, the normalized root mean square error (NRMSE) was used to determine when to stop the iterations, as shown in Eq. (8). The variation of NRMSE with the number of iterations is shown in Fig. 18 [Figure 18: see original paper]. It can be observed that the NRMSE reaches its optimal value at the 10th iteration. Therefore, the neutron energy spectrum after 10 iterations is selected as the final unfolded result of the GRAVEL method.

$$NRMSE(k) = \sqrt{\frac{\sum_{j=1}^{J} (\phi_j^{(k)} - \phi_j^{(real)})^2}{\sum_{j=1}^{J} (\phi_j^{(real)})^2}}$$

where ϕ_j^{(real)} is the intensity of the j-th bin of the true neutron spectrum.

After unfolding, the average relative deviation (ARD) and quality of the neutron spectrum (Q_s) are typically used to evaluate the accuracy of the unfolding method. These metrics are defined in Eq. (9) and Eq. (10), respectively:

$$ARD = \frac{\sum_{j=1}^{J} \left|\phi_j - \phi_j^{(real)}\right|}{\sum_{j=1}^{J} \phi_j^{(real)}}$$

$$Q_s = \sqrt{\frac{\sum_{j=1}^{J}(\phi_j - \phi_j^{(real)})^2}{\sum_{j=1}^{J}(\phi_j^{(real)})^2}}$$

After 10 iterations, the unfolding results of GRAVEL and the relative deviation (RD) of each bin obtained from the ARD expression are shown in Fig. 19 [Figure 19: see original paper] (a) and (b), respectively. The evaluation metrics ARD and Q__s are 0.31 and 0.19, respectively. As shown in Fig. 19, in the energy range of 6 MeV to 30 MeV, the unfolded spectrum obtained by the GRAVEL method matches the true spectrum well, and the RD values are also smaller. However, the unfolding performance degrades in the 30–200 MeV region, where RD values increase, leading to a higher ARD. The main reasons may be the deterioration of counting statistics in the high-energy region and the inaccuracy of Geant4's model cross section data in the high-energy region. Based on the Q__s value, the overall unfolding performance of GRAVEL remains satisfactory. These results demonstrate that the response matrix constructed entirely from experimental measurements, combined with the GRAVEL method, can effectively reconstruct the white neutron energy spectrum.

5. Conclusion

This study, based on the Back-n white neutron source at CSNS and combined with the Geant4 program, designed a 20 cm thick lead brick to effectively shield the accompanying gamma rays from the white neutron source. The experimentally measured and simulated neutron response energy spectra are both concentrated in the range of 0.1–300 MeV, with four significant and identical peak distributions within this range. The peak energies of the 1st, 3rd, and 4th peaks match well, with the absolute values of their relative deviations being less than 6%. However, the 2nd peak falls within the resonance energy region of lead, making it difficult to obtain an accurate peak energy. Due to the much higher elastic scattering cross section compared to the cross sections of (n, p), (n, d), (n, t), and (n, α) reactions in the low-energy neutron region, the intensity within the distribution range of the 1st and 2nd peaks (0.1–3 MeV) is lower than that of the 3rd and 4th peaks (5–64 MeV). This result demonstrates the effectiveness of the CLYC detector for high-energy neutron detection.

Furthermore, analysis of simulated and experimental results reveals that a fraction of incident neutrons undergo radiative capture and inelastic scattering with the CLYC, producing gamma rays. These gamma rays deposit energy in the CLYC detector, generating gamma pulse signals. This phenomenon is also reflected in neutron pulse signals. Specifically, when high-energy neutrons undergo nuclear reactions in the CLYC scintillator, secondary gamma rays and ions such as p, d, t, and α can be generated simultaneously, which renders the neutron pulse signals to include fast-decay components and makes the difference between neutron and gamma pulses less significant. As the GEE increases, this effect intensifies, reducing waveform distinctions between neutron and gamma pulses. These results indicate that for neutron detection, particularly for high-energy neutrons, relying solely on the R_{psd} from the charge comparison method or other pulse shape discrimination methods becomes increasingly inadequate for accurate neutron-gamma discrimination.

In addition, using the response matrix established from experimental measurements and the GRAVEL method, the white neutron energy spectrum in the range of 6–200 MeV was successfully unfolded. The unfolding achieved an ARD of 0.31 and a Q_s of 0.19. These results demonstrate the significant advantages of the CLYC detector in the field of wide-energy neutron measurements.

In future studies, the focus will be on signal reconstruction of pile-up pulses. This approach will eliminate the reliance on single-pulse events, allow flexible detector placement, and facilitate the rapid accumulation of sufficient pulse data in the study of wide-energy neutron spectrum measurements. It will also enhance the deployment of CLYC detectors in high-energy neutron experiments and space-based neutron detection. However, in the detection of high-energy neutrons, the original neutron pulse signal shape is affected by gamma signals, making the output neutron pulse shape of the detector more complex. This undoubtedly increases the difficulty of signal reconstruction, while also affirming the importance of this work. With accurate signal reconstruction as a foundation, research on neutron energy spectrum measurement and spectrum unfolding can further develop, improving the accuracy of spectrum measurement as well as the energy range and precision of spectrum unfolding. Meanwhile, in the study of the response of CLYC detectors to high-energy neutrons, by relying on precise neutron cross section data and the quenching factors of various reaction channels between neutrons of different energies and the main nuclides in CLYC scintillators, the detailed behavior of neutron interactions with CLYC detectors can be explored through Geant4 simulations. These advancements require the joint support of research fields such as nuclear cross-section measurement, crystal properties, and high-energy monochromatic neutron sources.

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