A novel 4π Gd-loaded liquid scintillator detection system postprint

Authors: YU Guo-Liang, DONG Lian-Jie, LONG Hou, LU Han-Lin, TANG Hong-Qin

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Abstract

With a Geant4 software package based on the Monte Carlo method, a multi-cell 4π detection system is designed, which consists of 40 Gadolinium-loaded liquid scintillation detectors. These detectors, associated with a fission chamber in its geometrical center, constitute a platform. This platform is mainly used for the measurement of a fissionable nucleus (n, 2n) reaction cross section. In order to properly determine the experimental set-up, we carry out a systematic numerical simulation using our model which is established by the Geant4 software package. This work provides rich and valuable reference data for experiments on the fissionable nucleus (n, 2n) cross section measurement in the future.

Full Text

Preamble

A Novel 4π Gd-Loaded Liquid Scintillator Detection System

YU Guo-Liang (于国梁)[1],†, DONG Lian-Jie (董连洁)[2], LONG Hou (侯龙)[3], LU Han-Lin (卢翰林)[3], and TANG Hong-Qin (唐洪庆)[3]

[1] Department of Mathematics and Physics, North China Electric Power University, Baoding 071003, China
[2] College of Science, Agriculture University of HeBei, Baoding 071001, China
[3] Institute of Nuclear Physics, China Institute of Atomic Energy, Beijing 102413, China

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Abstract: Using a Geant4 software package based on the Monte Carlo method, we have designed a multi-cell 4π detection system consisting of 40 gadolinium-loaded liquid scintillation detectors. These detectors, arranged around a fission chamber at the geometric center, form an integrated platform primarily intended for measuring (n, 2n) reaction cross sections of fissionable nuclei. To properly determine the experimental configuration, we performed systematic numerical simulations using our Geant4-based model. This work provides valuable reference data for future experimental measurements of (n, 2n) cross sections in fissionable nuclei.

Keywords: (n,2n) cross section, Geant4, Gadolinium-loaded liquid scintillator
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Introduction

The (n, 2n) nuclear reaction plays a crucial role in neutron multiplication within reactors, making accurate measurement of (n, 2n) cross sections for fissionable nuclei essential for nuclear energy utilization and nuclear technology development [1]. Researchers have employed various methods for this purpose. The activation technique is commonly used but requires the residual nucleus to have an appropriate half-life and demands high sample purity and intense incident neutron flux [2, 3]. The partial γ-ray method combines experimental measurements with theoretical nuclear reaction models, but its heavy reliance on theoretical modeling introduces significant uncertainties into the experimental results [2, 3].

In 1976, Frehaut et al. introduced a large gadolinium-loaded liquid scintillator with 80% neutron detection efficiency [4, 5]. The principle of this direct measurement method is illustrated in Fig. 1 [Figure 1: see original paper]. Two hemispherical containers hold the liquid scintillator, surrounded by a dozen photomultiplier tubes that collect signals from the Gd-loaded liquid scintillator. After collimation, monoenergetic neutrons irradiate samples positioned at the geometric center of the system. Neutrons from nuclear reactions are moderated within a few nanoseconds in the liquid scintillator, generating a neutron pulse (commonly called the fast signal). Since Gd isotopes, primarily ^{155}Gd and ^{157}Gd, have high thermal neutron capture cross sections, there is a high probability that neutrons will be captured by Gd. The excited Gd isotopes emit several cascade γ-rays with a total energy of approximately 8 MeV, and the pulse induced by these capture γ-rays is called the slow signal. Due to the high energy of this pulse, it can be easily discriminated from common background events such as γ-rays and scattered neutrons, which have energies below 3 MeV. This 8 MeV pulse thus indicates that a neutron was likely produced in the nuclear reaction, enabling precise determination of the number of neutrons generated.

This method does not depend on theoretical models and has no requirement regarding the half-life of the residual nucleus [4–7]. However, fission neutrons introduce significant deviations into the measured (n, 2n) cross sections for fissionable nuclei. Additionally, because the detection system has a large sensitive volume, background γ-rays produce a significant additive effect, resulting in high γ-ray background. Since each γ-ray penetrating the detection system can trigger responses in all photomultipliers, the total counting rate of the detection system becomes very high, leading to substantial dead time.

To address these limitations of the large Gd-loaded liquid scintillator technique, we have developed an improved 4π detection system consisting of 40 Gd-loaded liquid scintillator sub-detectors (Figs. 2 and 3). This multi-cell structure can effectively reduce the counting rate in individual detectors, thereby decreasing dead time. As discussed in Section III.A, this technique also provides certain (n, γ) discrimination capability, which effectively reduces the influence of γ-ray background on experimental results. To measure (n, 2n) cross sections in fissionable nuclei, samples are placed in a fast fission chamber located at the geometric center of the detection system. The pulse output from the fission chamber is used to exclude fission neutrons, greatly reducing the impact of fission events on experimental results. This platform can measure not only (n, 2n) and (n, 3n) cross sections but also neutron properties such as angular distribution and angular correlation, making the results presented here highly valuable for future neutron experiments.

II. Structures and Basic Performance of the Detection System

A. Structure of the Detection System

To achieve 4π solid angle coverage, adjacent detection units must connect seamlessly while maintaining approximately equal solid angles relative to the geometric center to ensure uniform detection efficiency. Based on Refs. [8, 9], we designed a detection system containing 40 detection units, numbered as shown in Fig. 2 [Figure 2: see original paper]. The system employs two types of detection units with pentagonal and hexagonal cross-sections, respectively. A cavity at the center of the detection system (Fig. 3 [Figure 3: see original paper]) accommodates the fast fission chamber. Additionally, to design the neutron beam tube, two detection units are removed from the system (Fig. 3). Each detection unit is an aluminum-shelled container approximately 3 mm thick, filled with standard liquid scintillator used in neutron experiments. The scintillator is mixed with Gd isotopes at a mass percentage concentration of 0.5%. The carbon-to-hydrogen ratio in the liquid scintillator is approximately 3:4, and its density is about 0.87 g/cm^3.

B. Performance of the Detection System

1. Response of the Detection System to the Neutron Source ^{252}Cf

The spontaneous fission neutron source ^{252}Cf is commonly used for efficiency calibration in neutron detection experiments. We simulated this neutron source positioned at the geometric center of the detection system and analyzed the system response using our Geant4 model. Partial γ-rays are emitted from excited Gd isotopes following neutron capture. Figure 4 [Figure 4: see original paper] shows the energy spectra deposited in both a single detection unit and the entire detection system. Since γ-rays primarily lose energy through the Compton effect in liquid scintillator [10], the γ-ray energy spectrum for a single detection unit forms a Compton plateau with an edge at approximately 8 MeV. The large sensitive volume of the detection system allows the energy of cascade γ-rays to be fully deposited through multiple Compton scatterings, forming a full-energy peak at 8 MeV. The neutron efficiency curve is shown in Fig. 5 [Figure 5: see original paper] (in Figs. 5 and 6, 'Th' denotes the threshold value of a single detection unit). Neutron efficiency is defined as $\epsilon = N_\gamma / N_n$, where N_γ is the total count in the γ-ray spectrum from neutron events and N_n is the total number of neutron events. In practice, detector thresholds are typically well below 1.0 MeV. Figure 5 shows that single-neutron efficiency exceeds 80% for all but the bottom curve and increases rapidly with detector thickness. When thickness exceeds 60 cm, the efficiency curve gradually plateaus. Figure 6 [Figure 6: see original paper] indicates that cavity radius variation has limited influence on two-neutron coincidence efficiency, except at a detector threshold of 2.0 MeV. To minimize overall system volume while maintaining high neutron efficiency, we preliminarily determined the detector thickness to be 60 cm. Furthermore, considering the placement of a fission chamber at the center, we selected a cavity radius of 30 cm.

2. Distribution of γ-Ray Energy Deposition in the Detection System
A ^{252}Cf neutron source placed at the geometric center emits neutrons toward detection unit 36 (Fig. 2). When a neutron is captured by Gd isotopes in unit 36, the deposited energy of capture γ-rays in each detection unit is recorded. Simulation results for the energy deposition distribution throughout the detection system are shown in Fig. 7 [Figure 7: see original paper]. The γ-rays deposit most of their energy in several adjacent units (36, 20, 21, 26, 11, and 27). These adjacent units, termed partial detection units, receive approximately 60% of the captured γ-ray energy. This occurs because, although the total γ-ray energy is approximately 8 MeV, the cascade typically includes several γ-rays of 1–2 MeV each, which have limited penetrating power and thus deposit most of their energy in nearby detection units. Consequently, partial detection units (comprising 6–7 adjacent units) respond to neutron-induced events. In contrast, background γ-rays are typically low-energy and deposit most of their energy in only one or two detection units, clearly distinguishing them from neutron events. Therefore, the number of fired sub-detection units can serve as a criterion for neutron events in future experiments, reducing environmental γ-ray background.

III. Numerical Simulations of the Experiment

A. Experimental Setup in Simulation

The fast fission chamber used in the detection system is a multi-target chamber that effectively reduces self-absorption of fission fragments without affecting the (n, 2n) reaction rate [11]. Aluminum alloy (duralumin) was selected as the structural material because aluminum nuclei have a relatively high (n, 2n) reaction threshold compared to other common metallic elements, and even when the (n, 2n) channel is open, its cross section remains small [11, 12]. The total mass thickness of the structural material is 2.7 g/cm^2. The fission chamber is positioned at the geometric center of the detection system with its axis aligned with the neutron beam tube. ^{235}U is plated on both sides of the electrodes, with a total sample mass thickness of approximately 100 mg/cm^2. The monoenergetic neutron source (14 MeV) is located 3.5 m from the geometric center of the detection system. Under these conditions, incident neutrons can induce various nuclear reactions.

B. Characteristic Time Distribution of Different Events

To properly configure the incident neutron pulse period, detector gate width, and timing relationship between the gate signal and neutron flux, we simulated the time distributions of different events that trigger responses in the liquid scintillator. These events include fast signals (elastic scattering neutrons, inelastic scattering neutrons, fission neutrons, and partial γ-rays from samples) and slow signals (capture γ-rays from excited Gd isotopes). In our simulation, the neutron emission time from the source is defined as zero. The time distributions of these events are shown in Fig. 8 [Figure 8: see original paper]. Prompt γ-rays cause the first detector response, followed by neutron events. The time distribution of these events is on the order of nanoseconds, while Figs. 8 and 9 show that the slow signal distribution is on the order of microseconds. Our simulation demonstrates that 99.5% of fast signals occur within 1 µs, with only about 0.2% of slow signals in this interval (blue curve in Fig. 8). Furthermore, 95% of slow signals occur within 30 µs, and over 99.7% within 50 µs.

The slow signal time distribution approximately equals that of neutron capture by Gd, which is the signal of interest for neutron counting. To avoid fast signal interference, the detector gate should open approximately 1 µs after neutron flux arrival. The detector gate width should be set to about 30 µs for neutron counting. The interval between consecutive neutron flux pulses is chosen as 60 µs, exceeding the neutron lifetime in the scintillator. In Refs. [4-6], Frehaut et al. used parameters of 1.2 µs, 30 µs, and 60 µs, respectively.

C. Estimation of Result Accuracy

With a total of 1.0×10^8 neutrons irradiating the samples in the fission chamber, we performed a systematic simulation of the experiment. Table 1 lists the numbers of different 2n events recorded by the detection system. These 2n events arise from uranium fission reactions, (n, 2n) reactions in uranium and aluminum, and accidental coincidences. The (n, 2n) reaction of nitrogen (the main component of air) must also be considered. If the chamber efficiency is approximately 90%, only 10% of 2n events from fission reactions (about 750 events in our simulation) cannot be excluded from the experimental results, reducing the influence of fission neutrons by an order of magnitude. When calculating 2n accidental coincidence events, the coincidence resolving time equals the detector gate width (30 µs), and the total neutron source intensity in the 4π direction is 10^8/s (an intensity of 10^8-10^9/s can be achieved at the accelerator of the China Institute of Atomic Energy).

The 2n background events from chamber structural materials (aluminum) and atmospheric nitrogen can be subtracted by measuring with the neutron beam irradiating the fission chamber without samples under identical conditions. Simulation also shows that only 10% of accidental coincidences are sample-related [13], meaning 90% can be subtracted using the same method. Therefore, the counting uncertainty of 2n events from aluminum, nitrogen, and accidental coincidences can be combined, and the net count uncertainty can be estimated as $\sqrt{\sigma_t^2 + \sigma_f^2 + \sigma_b^2} = 135$, where $\sigma_t = 117.38$, $\sigma_b = 60.15$, and $\sigma_f = 27.36$ as listed in Table 1. The relative uncertainty is 1.43%, where $N_{net} = 9411$ represents the signal of interest.

Certain factors such as accidental coincidences with cosmic rays and γ-ray spectrum energy resolution have not yet been considered. Sample content calculations may have some deviation, and neutron flux variations can also introduce errors. Thus, (n, 2n) cross section measurements will be influenced by many factors beyond those considered here, and the actual experimental uncertainty will be larger than our current estimate. Nevertheless, our simulation results remain important for the future design and optimization of experimental conditions.

IV. Conclusion

Based on the experimental setup of Frehaut et al., we have designed a novel 4π multi-cell Gd-loaded liquid scintillator detection system for measuring (n, 2n) nuclear reaction cross sections. This system addresses the weaknesses of the original design used by Frehaut et al. Through systematic simulation, we have preliminarily determined the detector thickness, inner radius of the detection system, neutron source pulse period, detector gate width, and timing relationship between gate signal and neutron flux. Finally, we have roughly estimated the accuracy of experimental results under our proposed setup.

Further work remains to be done. For the γ-ray spectrum, our simulation currently considers only energy loss processes in the detector, not fluorescence photon collection. Including this process would result in lower energy resolution, which significantly influences efficiency. Therefore, the photon collection process should be incorporated into our Geant4 code to better match experimental conditions. Additionally, to reduce the influence of large-angle scattered neutrons, cosmic rays, and environmental γ-rays, we will devote considerable effort to designing the neutron beam tube and shielding layer around the detection system, including selection of shielding materials, calculation of shielding thickness, and determination of neutron tube diameter.

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