

A long ^3He proportional counter array for the study of β -delayed neutron emission probability at BRIF

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Abstract

The β -delayed neutron emission probability (P_n) is an indispensable quantity to describe the decay strength of very neutron-rich nuclei and the rapid neutron capture process in nuclear astrophysics. A Long Helium-3 Neutron Array (LHENA), is developed at the Beijing Rare Isotope Facility (BRIF) to initiate P_n measurements using Isotope Separated On Line (ISOL) pulsed beams. LHENA is designed to work in conjunction with a tape driver and different detectors, so that β particles, β -delayed neutrons and γ rays emitted from the implanted nuclei can be measured simultaneously in periodical mode. LHENA consists of 21 long ^3He proportional counters embedded in a polyethylene-made moderator in two-ring structure, which allows for a flat neutron detection efficiency up to 3 MeV according to our Geant4 simulation. The detection efficiency has been experimentally determined to be $16.4(\pm 0.4)\%$ using the $^{51}\text{V}(p,n)^{51}\text{Cr}$ reaction for neutron energies in the 120-700 keV range. A good flatness in neutron detection efficiency and very low background of LHENA are verified, which lay a solid foundation for the first P_n measurement using very neutron-rich Rb isotopes at BRIF.

Full Text

Preamble

A Long ^3He Proportional Counter Array for the Study of β -Delayed Neutron Emission Probability at BRIF

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The β -delayed neutron emission probability (P_n) is an indispensable quantity for describing the decay strength of very neutron-rich nuclei and the rapid neutron capture process in nuclear astrophysics. A Long Helium-3 Neutron Array (LHENA) has been developed at the Beijing Rare Isotope Facility (BRIF) to initiate P_n measurements using Isotope Separated On Line (ISOL) pulsed beams. LHENA is designed to work in conjunction with a tape driver and different detectors, enabling simultaneous measurement of β particles, β -delayed neutrons, and γ rays emitted from implanted nuclei in periodic mode. LHENA consists of 21 long ^3He proportional counters embedded in a polyethylene moderator in a two-ring structure, which allows for a flat neutron detection efficiency up to 3 MeV according to our Geant4 simulation. The detection efficiency has been experimentally determined to be $16.4(\pm 0.4)\%$ using the $^{51}\text{V}(p,n)^{51}\text{Cr}$ reaction for neutron energies in the 120–700 keV range.

The good flatness in neutron detection efficiency and very low background of LHENA have been verified, laying a solid foundation for the first P_n measurement using very neutron-rich Rb isotopes at BRIF.

Keywords: ^3He proportional counter, Long Helium-3 neutron array, β -delayed neutron emission probability, Beijing Rare Isotope Facility (BRIF)

INTRODUCTION

β -delayed neutron emission was first observed in 1939 accompanying the neutron-induced fission of uranium and thorium isotopes [?]. These neutrons are emitted in the β^- decay process of neutron-rich fission fragments when the β decay energy (Q_β) is larger than the neutron separation energy (S_n) of the daughter nuclei, i.e., $Q_{\beta 1n} = Q_\beta - S_n > 0$. The β -delayed neutron emission probability P_n is defined as the fraction of delayed neutrons in the decay [?], which demonstrates the fraction of β strength above S_n . Although the average number of β -delayed neutrons per fission ($\langle \mu_d \rangle$) is very small for fission systems in nuclear energy applications, β -delayed neutrons play a key role in safely

controlling the prompt chain reaction at criticality in nuclear power plants [?]. β -delayed neutron groups from different fission fragments were identified [4–9], and a six-group fitting parameter set was established by Keepin and co-workers in 1957 [?]. To obtain $\langle\mu_d\rangle$ for different fission systems, the summing method may also be applied besides direct measurement, by using the fission cumulative yield Y and the P_n of each fragment, i.e., $\langle\mu_d\rangle = \sum_i Y_i \times P_{ni}$ [?]. To date, large deviations exist between results from these two methods, even for the thermal neutron-induced fission of ^{235}U , which is partly due to the large deviations in available P_n data from different measurements.

P_n increases quickly with increasing $Q_{\beta 1n}$ values, and β -delayed multiple neutron emission channels become probable if $Q_{\beta xn} > 0$ for very neutron-rich nuclei [?]. In this scenario, the β decay property and nuclear structure are largely accessible only through β -delayed neutrons [?]. In the rapid neutron capture process (r-process) of nuclear astrophysics [?, ?], the nucleosynthesis path runs close to the neutron drip line in the nuclide chart. A large number of β -delayed neutron emitters are involved, and delayed neutron emission modulates the heavy element abundance curve by altering the decay route [?, ?] and supplying a fresh neutron source [?]. Precise experimental data from delayed neutron emission represent a key input to r-process model calculations.

Thousands of neutron-rich isotopes are predicted to be β -delayed one- or multi-neutron emitters according to the present Atomic Mass Evaluation in 2020 (AME2020) [?]. The quality of existing experimental data is not sufficient for various technical and scientific applications. It is therefore urgent to perform new measurements with leading-edge neutron detector arrays and high-quality neutron-rich beams, which have become available at BRIF [?] recently and will be available at the High Intensity heavy-ion Accelerator Facility (HIAF) [?, ?] in the near future. We report in this paper the installation of LHENA at BRIF for the study of β -delayed neutron emission probability. The LHENA design and overall P_n detection system are described in Section 2. The determination of LHENA detection efficiency is detailed in Section 3. Results are presented in Section 4, followed by a short discussion and summary in Section 5.

II. LHENA SETUP

A. Method

By definition, P_n describes the fraction of delayed neutrons in the β decay; therefore, it can be simply deduced by the following formula:

$$P_n = \frac{N_n/\epsilon_n}{N_\beta/\epsilon_\beta}$$

where N_n and N_β are the counts of detected neutrons and β particles, respectively; ϵ_n and ϵ_β refer to the detection efficiencies of neutrons and β particles, respectively. If the half-life of the nuclide is short enough compared to the very

weak beam intensity, the counts of β particles could be approximated by that of the implantation ion, which is useful in studying P_n of very rare isotopes produced at projectile fragmentation facilities like HIAF.

The β -delayed neutrons could be measured with the Time-Of-Flight (TOF) method in principle to obtain their kinematic energy information. However, this is only feasible for very few isotopes that can be produced with high intensity. In most cases, a proportional counter array with very high detection efficiency must be employed to compensate for the very low beam intensity of very neutron-rich nuclei. For such a purpose, a neutron detection system based on the ^3He gas proportional counter has been very popular for its high sensitivity, low intrinsic activity, and excellent durability [23–27]. It works through the thermal neutron capture reaction $n + ^3\text{He} \rightarrow ^1\text{H} + ^3\text{H} + 765\text{ keV}$ at the maximum cross section of about 5330 barn. Such a high-efficiency ^3He neutron counter array is free from cross-talk of multiple scattering, thus allowing for neutron multiplicity measurements [?].

Normally, P_n measurement is performed in the β -n coincidence mode [?], which can be expressed as:

$$P_n = \frac{N_{\beta-n}}{N_\beta \cdot \epsilon_n \cdot \epsilon'_\beta}$$

where $N_{\beta-n}$ is the counts of neutrons detected in coincidence with β particles, and ϵ'_β represents the β detection efficiency with energies corresponding to the neutron emission. When ϵ'_β equals ϵ_β , Equation 5 is simplified as:

$$P_n = \frac{N_{\beta-n}}{N_\beta \cdot \epsilon_n}$$

By using β -n coincidence, one avoids the precise determination of ϵ_β . When the neutron counting rate is low, this method has the advantage of largely improving the signal-to-background ratio [?].

Since the measured β particles and neutrons usually do not have clear energy information, it is necessary to combine HPGe detectors in the setup to enable characteristic γ -ray measurement for nuclide identification. Furthermore, the β detection efficiency could be evaluated through the γ -ray intensity ratio with and without coincidence with β particles [?]. The neutron detection efficiency could be evaluated in a similar way with the help of the characteristic γ -ray intensity.

B. LHENA Description

The schematic of the long ^3He proportional counter array LHENA is shown in Figure 1: see original paper. LHENA consists of 21 long ^3He proportional

counters manufactured by GE-Reuter Stokes. The active length of the ^3He proportional counter is 800.0(31.5) mm with a diameter of 25.4(0.8) mm. Each counter is filled with ^3He gas at 4 bar pressure, sealed with stainless steel housings. When a thermal neutron is captured in the sensitive volume of the ^3He gas, the energy released from the reaction is converted into the kinetic energies of the triton (191 keV) and the proton (574 keV). A full-energy peak of 765 keV is observed if both particles fully deposit their energies within the ^3He gas volume. However, due to the finite detector volume, either the proton or triton may hit the sealing wall and lose part of its energy. This phenomenon is known as the wall effect [?]. To reduce the wall effect and improve the peak-to-plateau ratio in the energy spectrum, argon gas at 1.5 bar pressure is mixed in the ^3He proportional counter for its strong stopping power.

The ^3He proportional counters are embedded in a polyethylene-made neutron moderator. A central borehole with a diameter of 10 cm is created in the moderator to accommodate the beam line and target chamber. This compact configuration allows for nearly 4π solid angle coverage around the ion implantation position. The ^3He proportional counters are distributed in two concentric rings: three proportional counters are installed in the inner ring at 7.6 cm radius, and 18 proportional counters in the outer ring at 18 cm radius. This configuration enables a flat neutron detection efficiency up to 2.5 MeV according to our Geant4 simulation. The polyethylene moderator is surrounded by a 5 cm-thick 7% boronated polyethylene layer to shield against environmental neutrons. The overall dimensions of LHENA are 70 cm \times 70 cm \times 100 cm.

As shown in Figure 1: see original paper, the pulsed neutron-rich ISOL beam from BRIF is implanted in a tape situated at the center of LHENA. The β particles are measured with a silicon detector close to the tape in the vacuum chamber. The β -delayed neutrons and γ rays are simultaneously measured with LHENA and an HPGe detector, respectively. To remove long-lived isobaric contaminants and daughter nuclei, the P_n measurement will be performed in a periodic mode, with a preset period of radioactivity growth-in and decay according to the half-life of the nucleus under study. After each cycle, the tape is moved to a new position for the next-cycle ion rate as shown in [Figure 2: see original paper]. The neutron counting rate for a single ^3He proportional counter should be kept low, below 120/s for example, to maintain a marginal pile-up effect (less than 0.1%).

For P_n measurement, it is highly desirable that the detection efficiency is flat over a wide neutron energy range. Therefore, the detection efficiency and energy dependence of LHENA must be known clearly. For this purpose, the $^{51}\text{V}(p,n)^{51}\text{Cr}$ reaction is a widely-used reaction to calibrate neutron arrays [23, 30–34] like LHENA. The neutron energy and intensity produced by the reaction vary slowly with angle, making it an ideal quasi-monochromatic and isotropic neutron source. In addition, ^{51}Cr has a half-life of 27.7 days and emits a 320 keV characteristic γ ray. The number of neutrons produced by the reaction could be determined by the activation method with high accuracy,

without having to rely on reaction cross-section data. However, its application is limited to $E_p = 2330$ keV, above which an additional neutron channel opens corresponding to the first excited state of ^{51}Cr at $E_x = 749$ keV.

The experiment was carried out at the nuclear physics experiment (NPE) terminal of the 3-MV tandetron accelerator [?, ?] at Sichuan University. The ^{51}V target used in the experiment was made by evaporating natural vanadium onto 1 mm thick tantalum with a diameter of 15 mm. The nominal thickness of ^{51}V is $100 \mu\text{g}/\text{cm}^2$, corresponding to an 8–10 keV proton energy loss at $E_p = 1700$ – 2300 keV. The 1 mm thick tantalum substrate ensures the mechanical strength required for vacuum sealing. The experimental setup consists of stainless steel pipes with an outer diameter of 90 mm and a wall thickness of 3 mm, as shown in [Figure 3: see original paper]. Inside, there is a coaxial copper cryotrap pipe with an outer diameter of 60 mm and a wall thickness of 2 mm, which was cooled by liquid nitrogen to prevent carbon buildup on the target surface. At the end of the cryotrap pipe, a 5 mm diameter tantalum collimator was installed. The negative voltage ring, with a diameter of 15 mm and a thickness of 2.5 mm, was fixed to the collimator by insulated screws. A voltage of -600 V was applied during the experiment.

III. EXPERIMENT FOR LHENA EFFICIENCY CALIBRATION

A typical neutron pulse-height spectrum measured by a ^3He proportional counter with an Am-Be neutron source is shown in [Figure 2: see original paper]. Signals from γ rays, electronic noise, and β particles can be clearly distinguished from neutron signals in the spectrum. A test pulse of 1 Hz frequency from a precise pulse generator is also recorded to monitor system operation and dead time.

The neutron signal from the ^3He proportional counter exhibits a long pulse feature, with a duration of about $500 \mu\text{s}$ and a rise time of about $5 \mu\text{s}$. This could lead to significant pulse pile-up effects in high counting-rate experiments, which hinders precise deduction of P_n values. The relationship between the pile-up rate and counting rate is systematically studied by varying the position of the Am-Be neutron source in LHENA. The pile-up rate increases linearly with the neutron counting rate.

The ^3He proportional counters were operated at a bias voltage of 1200 V, supplied by an ORTEC 660 high-voltage power supply. The signals from each proportional counter were processed by a CAEN A1422 charge-sensitive preamplifier with a gain of 45 mV/MeV. The amplified signals were then acquired by the CAEN R5560 digital acquisition system, which has a sampling rate of 125 MS/s and a resolution of 14 bits. The data were transmitted via USB to a computer for storage. The accumulated charge on the target was measured using an ORTEC 439 digital current integrator.

Since the inner ^3He proportional counters have much higher detection efficiency for low-energy neutrons, which results in significantly higher count rates compared to the outer ones, a CAEN DT5740D desktop digitizer system was used in combination with CoMPASS software to enable real-time monitoring of counting rates. Two inner-ring ^3He proportional counters were monitored to avoid heavy pile-ups, and one outer-ring ^3He counter was monitored for sufficient statistics. During the experiment, the counting rate for a single ^3He proportional counter in the inner ring was kept below 120 counts/s, and the overall counting rate of the LHENA system was below 500 counts/s. The dead time of the DAQ system is completely negligible. We used the same ^{51}V target for beam tuning and replaced it with a new ^{51}V target for measurements. In total, we made measurements at $E_p = 1700, 1850, 2000, 2150, \text{ and } 2300$ keV, corresponding to $E_n = 120, 265, 410, 550, \text{ and } 695$ keV, respectively.

The number of neutrons emitted from the $^{51}\text{V}(p,n)^{51}\text{Cr}$ reaction is equivalent to the number of radioactive ^{51}Cr nuclei, which decay through electron capture with a half-life of 27.7 days. The total number of emitted neutrons at each energy was therefore determined through measurement of the 320 keV γ ray from the activated targets [?, ?, ?]. The number of reactions (N_R) occurring during the activation time (t_i) can be obtained using the following formula:

$$N_R = \frac{N_\gamma}{\epsilon_{320} \cdot \eta_{320}} \cdot \frac{1 - e^{-\lambda t_c}}{\lambda \cdot t_i} \cdot \frac{1}{1 - e^{-\lambda t_i}}$$

where N_γ is the net counts of the 320 keV γ rays, t_c is the counting time, t_w is the waiting time elapsed between the end of irradiation and the start of γ counting, and λ is the decay constant of ^{51}Cr .

The neutron detection efficiency ϵ_n is simply calculated by:

$$\epsilon_n = \frac{N_n}{N_R}$$

where N_n is the counts of neutrons recorded by LHENA.

The activated targets were measured using the low-background Gamma spectrometer for Nuclear Activation Studies (GNAS) [?] at CIAE, which consists of a well-type HPGe detector surrounded by optimized multi-layer shielding. GNAS has a near 4π geometry for small radioactive samples, thus being very efficient for radiochemical analysis and low-level γ -ray spectroscopy. A ^{51}V target was irradiated with a high-flux proton beam in the experiment, and the activity of ^{51}Cr was calibrated using a standard γ spectrometer [?] at CIAE. The absolute efficiency of GNAS was then determined to be 34.2%(1.2%) for the 320 keV γ ray.

A typical γ spectrum of the ^{51}Cr sample measured by GNAS is shown in [Figure 4: see original paper].

The background neutron counting rate in the experimental hall is $0.138(0.001)/s$, which is negligible compared to about 500 neutrons/s detected by LHENA. The neutron background originating from proton bombardment of the tantalum backing was also evaluated by comparing normalized neutron yields under beam focusing and defocusing conditions at $E_p = 1.7$ MeV. The introduced uncertainty was estimated to be less than 0.5%.

IV. RESULTS

LHENA position sensitivity along the beam axis was studied using the $^{51}\text{V}(p,n)^{51}\text{Cr}$ reaction at $E_p = 2$ MeV at thirteen positions with $Z = 0, \pm 3, \pm 6, \pm 10, \pm 15, \pm 20$, and ± 25 cm, respectively. The position with $Z = 0$ corresponds to the target aligned at the center of LHENA. Positive Z -values indicate forward displacement of LHENA along the beam direction, while negative values represent backward displacements. The relative efficiencies at different positions were normalized to the value at $Z = 0$ using the integrated incident beam currents on the target. The experimental and simulation data are normalized to 1 at $Z = 0$, as shown in [Figure 5: see original paper]. Excellent agreement is shown between experimental data and simulations. Displacement of the neutron source within ± 10 cm has negligible impact on the relative efficiency, primarily benefiting from the extended sensitive region of the ^3He proportional counters. About 5% efficiency asymmetry was observed in the ± 10 to ± 25 cm displacement range, which might be attributed to structural asymmetry of the polyethylene moderator and/or the ^3He proportional counters. This small asymmetry does not impact P_n experiments, where radioactivity implantation occurs in the central region of LHENA and should not exceed the ± 10 cm range. Furthermore, benchmark measurements will be carried out with the same LHENA geometry for nuclei with well-known P_n values.

The experimental neutron detection efficiency of LHENA is listed in and shown in [Figure 6: see original paper], in comparison with Geant4 simulations. The experimental data show an average efficiency of $16.4(\pm 0.4)\%$ across five measurement points, verifying good flatness in neutron detection efficiency.

The Geant4 simulation [cite{40–42}] was performed independently for each ring as shown in [Figure 6: see original paper], which agrees reasonably well with the trend of the experimental data. The agreement is slightly worse for the outer ring at the lowest neutron energies. According to Geant4 simulation, the total detection efficiency of LHENA remains nearly constant within the 0–3 MeV energy range, with only a marginal decrease of 1.2% at 5 MeV. The good flatness of LHENA detection efficiency over a wide neutron energy range makes P_n measurement feasible for a large number of neutron-rich isotopes at BRIF.

As shown in [Figure 6: see original paper], our Geant4 simulated ϵ_n efficiencies exhibit a systematic overestimation of approximately 6%. To compare the agreement of trends, a scale factor was applied from the ratio of experimental-to-

simulated efficiencies at $E_n = 550$ keV. Different scaling factors were employed for the inner ring and total efficiencies. The scaled outer ring efficiency is the difference between scaled total and inner ring efficiencies. The simulation results follow the energy dependence of the measured data very well after applying the scaling factors.

V. DISCUSSION AND SUMMARY

Neutron detector arrays based on ^3He proportional counters are widely used in many applications requiring high-efficiency neutron detection. Although a very large neutron energy range may be involved (for example, in γ - or charged-particle-induced neutron generation reaction studies), a flat detection efficiency curve is a major advantage for obtaining absolute neutron yields regardless of their energies. Since mono-energetic neutron sources are very scarce, the determination of detection efficiency for such neutron detectors relies largely on simulations, which is one of the major sources of systematic errors in obtained data.

As shown in this work, the simulated results exhibit notable discrepancies compared to experimental efficiency data. Such overestimation has been widely observed for similar neutron arrays [?, ?, ?], demonstrating the importance of experimental verification. The reason for the overestimation of detection efficiency in simulation is not clear. Different corrections have been arbitrarily adopted in simulations, including altering the geometry and density of the polyethylene moderator and the pressure of the ^3He counter [?]. The possibility of mixing a small amount of boron in the polyethylene moderator is also discussed in Ref. [?]. We have tested these presumptions in simulation, but none seem to provide a reasonable and conclusive solution. In fact, the neutron scattering cross-sections employed in simulations contain uncertainties, particularly in modeling molecular effects during neutron moderation within polyethylene and the inherent stochastic nature of neutron scattering. A larger deviation is indeed observed for simulation of outer ring efficiency with longer neutron travel paths. Whether or not this discrepancy stems from uncertainties in the neutron scattering cross-sections used in simulations, it is important to expand experimental verification to higher neutron energy regions.

The activation technique in Equation (2) holds under the assumption that beam intensity remains constant. However, during actual experiments, beam intensity fluctuates over time and occasionally needs to be intentionally increased to compensate for lower counting rates. Since the variation of neutron counting rate is equivalent to that of the ^{51}Cr yield, we used the time-dependent behavior of the neutron counting rate to evaluate the introduced uncertainty. By comparing the total ^{51}Cr yield based on the average neutron counting rate and considering beam intensity fluctuation, the difference was found to be approximately 0.007%, indicating this error is negligible. This result is primarily attributed to the fact that the half-life of ^{51}Cr is much longer than the irradiation duration.

In summary, a long ^3He neutron proportional counter array has been developed for the study of β -delayed neutron emission probability at BRIF. The design and performance of LHENA have largely relied on extensive Monte Carlo simulations. To verify the overall design, the absolute detection efficiency of LHENA has been determined using the $^{51}\text{V}(p,n)^{51}\text{Cr}$ reaction for neutron energies in the 120–700 keV range. The results show that after applying a scaling factor, the simulated efficiency is in good agreement with the energy-dependent trend of the experimental efficiency. The good flatness of LHENA detection efficiency over a large neutron energy range makes it universally applicable for P_n measurements at BRIF and neutron source reactions in nuclear astrophysics at JUNA as well.

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