Improved fission neutron energy discrimination with ^4He detectors through pulse filtering

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Abstract

This paper presents experimental and computational techniques implemented for ^4He gas scintillation detectors for induced fission neutron detection. Fission neutrons are produced when natural uranium samples are actively interrogated by 2.45 MeV deuterium-deuterium fusion reaction neutrons. Fission neutrons of energies greater than 2.45 MeV can be distinguished by their different scintillation pulse height spectra since ^4He detectors retain incident fast neutron energy information. To enable the preferential detection of fast neutrons up to 10 MeV and suppress low-energy event counts, the detector photomultiplier gain is lowered and trigger threshold is increased. Pile-up and other unreliable events due to the interrogating neutron flux and background radiation are filtered out prior to the evaluation of pulse height spectra. With these problem-specific calibrations and data processing, the ^4He detector's accuracy at discriminating fission neutrons up to 10 MeV is improved and verified with ^{252}Cf spontaneous fission neutrons. Given the ^4He detector's ability to differentiate fast neutron sources, this proof-of-concept active-interrogation measurement demonstrates the potential of special nuclear materials detection using a ^4He fast neutron detection system.

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1. Introduction

The recent global shortage of ^3He has motivated development of fast neutron detectors based on alternative materials. In particular, ^4He gas is naturally abundant and can act as both the neutron detection medium and scintillator in
20 a detector. In this work, we focus on the application of ^4He fast neutron gas scintillation detectors, developed by Arktis Radiation Detectors Limited [1], to the detection of a relatively weak fission neutron signal in the presence of a strong background. This condition is expected in realistic applications such as the active interrogation of special nuclear materials and spent nuclear fuel
25 source term determination [2–6]. Based on ^4He and neutron elastic scattering at fast neutron energies, ^4He detectors can detect fast neutrons without any neutron moderating materials. The ability of ^4He detectors to retain incident neutron energy information is a crucial advantage since the energy of neutrons from (induced or spontaneous) fission or the (α, n) reaction can range from a few
30 hundred keV to 10 MeV. Previous works have shown, in both simulations and preliminary experiments, the detector’s ability to differentiate induced fission neutrons from the interrogating neutrons [7–10]. However, it has also been observed that the experimental measurements can be affected by pulse pile-up and false neutron counts in a strong background. Although ^4He detectors are
35 most sensitive to 1 to 2 MeV neutrons [11], in this work we show how they can be used to effectively detect much higher energy neutrons, despite a large background flux of interrogating neutrons, by using problem-specific calibrations and pulse processing techniques.

The compact ^4He fast neutron detector consists of a stainless steel cylindrical
40 chamber filled with pressurized ^4He gas close to 150 bar and two Hamamatsu-R580 photomultiplier tubes (PMTs) installed at the two ends of the chamber (see Figure 1). The inner wall of the chamber is lined with a wavelength shifter

to convert ultra-violet scintillation to the optical wavelengths for the PMTs. A coincidence setting requires that both PMTs detect scintillations within a 32 ns window, so as to suppress the recording of PMT noise signals.

Figure 1: ^4He fast neutron detector is manufactured by Arktis Radiation Detectors Ltd.

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Figure 2: $^4\text{He}(\text{n,elastic})$ cross section compared to the Watt fission probability spectrum above 200 keV.

The cross section of ^4He (n,elastic) has a resonance peak at approximately 1.125 MeV, which almost matches with the peak fission neutron energy spectrum as shown in Figure 2. Through elastic scattering, an incident neutron transfers up to 64% of its energy to a recoil α particle depending on the elastic scattering angle distribution [11]. The recoil α particle subsequently ionizes and excites

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other ^4He atoms along its path, resulting in two excited molecular states of ^4He , namely the singlet and triplet states [12]. The distinct de-excitation time-scales of these two states are seen in Figure 3 as the fast and slow component respectively. In Figure 3b, the overall lower light yield for a gamma event is due to the low electron charge density of ^4He and the low ionization density of recoiling Compton electrons before they are absorbed by the detector inner wall [1]. ^4He detectors are considered excellent in rejecting gamma events and the difference in the relative strength of the two components for neutrons and gammas enables the effective pulse shape discrimination [13].

Figure 3: Typical digitized PMT outputs of neutron and gamma events. Fast component lasts approximately 50 ns, followed immediately by the slow component till the end of the $4.27\mu\text{s}$ event window.

In a proof-of-concept experiment, a deuterium-deuterium (D-D) fusion-based neutron generator is used for the active interrogation of natural uranium. The

D-D reaction generates monoenergetic neutrons of 2.45 MeV which can induce fission neutrons in fissionable materials. The maximum deposited energy from the monoenergetic neutron generator is 64% of 2.45 MeV or 1.57 MeV. Subsequently, its maximum scintillation pulse height value can be used to discriminate fission neutrons which can deposit energies above 1.57 MeV and generate pulse heights greater than those of D-D neutrons.

However, several factors affect the determination of the maximum D-D pulse height value. During the operation of the neutron generator, the ^4He detector is also subjected to background radiation such as Bremsstrahlung X-rays and neutrons/gammas scattered back from the HDPE-covered walls of the experiment room. Pulse pile-up can artificially increase the calculated pulse height values, such that the combined scintillation light from several lower energy events can be falsely attributed to the deposited energy of a higher energy neutron. Conversely, due to pulse saturation, a suppressed value of the maximum pulse height can lead to underestimation of the incident neutron energy.

The goal of the current work is to improve the ability of ^4He detectors to differentiate fission neutrons from generator-emitted neutrons with problem-specific pulse height calibration and filtering. Section 2 provides the details of the calibration steps to address the aforementioned causes of signal defects. Section 3 presents the measurements of the active interrogation of natural uranium samples and the improvement on the accuracy of the fission neutron energy discrimination in the pulse height spectrum.

2. ^4He detector calibration

Under normal operation, the highest sensitivity and efficiency of the ^4He detector is in the 1 to 2 MeV range, given the location of the ^4He (n,elastic) cross section peak in Figure 2. In this work, the neutron energy of interest is above the monoenergetic 2.45 MeV from the D-D neutron generator, and up to 10 MeV for fission neutrons. During generator operation, the generator flux and background radiation make up the majority of the detected signal compared

to the induced fission neutron signal. Therefore, problem-specific calibrations have to be implemented to increase the probability of recording a high energy neutron event by suppressing the trigger rate of low energy events from the large interrogating neutron flux and background radiation. Post-processing of ⁴He detector output data can also further distill the fission neutron signals by filtering out pile-up and unreliable events associated with a high count-rate background.

2.1. Gain and threshold adjustment

Each ⁴He detector has two Hamamatsu-R580 PMTs to collect scintillation light caused by incident particle interactions. The PMT output signals are sampled at 120 MHz by a custom-built (by Arktis) data acquisition system (DAQ) and the digitized scintillation waveforms are summed to produce the pulse height of each event. The gain of the DAQ readout electronics can also be varied such that the amplified signals of high energy events are still within the dynamic range of the DAQ. To calibrate the detector system, a 0.076 μ Ci ⁶⁰Co gamma source was placed at the center position on top of the ⁴He detector active volume. At different PMT voltages, 10^4 events were recorded and the ⁴He detector DAQ calculated the corresponding gain calibration factors [12]. To match the gain of the two PMTs of the same ⁴He detector, the PMT voltage values can be obtained from the linearly fitted relations between voltage and gain in Figure 4. Since the main interest of this work is to detect energetic events, the PMT voltages were lowered from the default settings to prevent signals from exceeding the DAQ dynamic range. Furthermore, a DAQ trigger threshold parameter was set higher than its default value, so that events with small light yield in the fast component would not be recorded.

2.2. Event filter

For the accurate estimation of the deposited energy of the incident neutron, it is necessary to filter out events whose pulse heights do not in fact represent

Figure 4: Linear fits for the relationship between the PMT voltages and the overall gain values of the DAQ electronics.

the actual energy deposition of a single high energy neutron scattering event.

120 Spurious triggering and pile-up are two primary causes of unusable events.

Normally the coincident fast components of the two PMTs trigger the DAQ to record the event waveform, including a $0.5 \mu\text{s}$ pre-trigger to establish the event baseline. A spurious trigger is when the coincidence trigger requirement is satisfied by chance as shown in Figure 5. The true fast component scintillation
 125 peaks and the subsequent slow component scintillation signals occur later in the same event window. A spurious trigger can cause both an erroneous calculation of the integrated fast component value and a loss of slow component data. Detection of spurious trigger events is accomplished by comparing the largest slow component pulse amplitude with the fast component peak. The former
 130 should not exceed the latter, otherwise the event is filtered out and not used in the pulse height spectrum calculation.

The PMT signal waveform has a default duration of $4.27 \mu\text{s}$ (or 512 sample points at 120 MHz) to capture the full range of the slow component on the order

Figure 5: A spurious trigger occurs when the PMT coincidence requirement is satisfied by the PMT noise signals. A true but incomplete neutron event is recorded in the time window. These event waveforms are unusable.

of microseconds. For high energy events, it is possible that the slow component
 135 scintillations of another event are captured upon a separate event trigger, as
 shown in Figure 6. It is not only difficult to reconstruct the corresponding pre-
 trigger baseline values, but the slow and fast components of the two consecutive
 events also overlap in a single event window. Hence such events are filtered from
 the calculation of the pulse height spectrum.

140 In a high count-rate environment, pile-up events are those that occur within
 a single event window. In Figure 7, a second event is captured in the same event
 window along with the first triggered event. Such event waveforms are unusable
 for the calculation of the pulse height of supposedly separate events, and must be
 discarded. To differentiate pile-up events from those genuine waveforms which
 145 are caused by high energy neutrons and hence have large slow components, a
 pile-up filter is made to compare the maximum amplitude of the slow compo-
 nent trace to a pre-defined threshold. The value of the threshold is chosen by
 examining the ratios between the maximum amplitude of the slow component
 and the fast component peak (as labeled in Figure 7) of a ^{252}Cf source. Among
 150 about 2.4×10^4 events recorded with the ^4He detector, the spurious trigger filter

Figure 6: Scintillation from a presumably large energy transfer “overflows” into the next interaction event.

first removed 0.8% of the total events. The rest of the events were separated into neutrons (81%) and gammas (19%) with the charge integration pulse shape discrimination method as shown in Figure 8. The ratio between the maximum amplitude of slow component and the fast component peak was calculated for each event and the distributions of the ratios are plotted in Figure 9. As expected, gamma events have lower ratio values due to low light yields in the slow component. It was calculated that 99.2% of the neutron events had a ratio value less than 0.6. Given that ^{252}Cf spontaneous fission neutrons have a wide range of fast energies, it is assumed that the distributions shown in Figure 9 conservatively represent other fast neutron sources such as the D-D neutrons and induced fission neutrons. Therefore, the maximum amplitude of the slow component is required to be less than 0.6 times the fast component peak; otherwise the event is filtered out by the pile-up rejection algorithm in this work.

3. Measurement results

The ^4He detector was used to measure induced fission neutrons in the background of the interrogating neutrons. Given the ability of the ^4He detector to retain incident neutron energy information, the unambiguous detection of

Figure 7: Within the time window of a neutron event, a second event is also recorded.

Figure 8: ^{252}Cf neutron events are found above the pulse shape discrimination (PSD) line since neutron events have larger integrated slow component as shown in Figure 3. Gamma events are below the PSD line.

fission neutrons, which have higher energies than the interrogating neutrons, shall demonstrate the potential of ^4He detectors for special nuclear materials detection.

Figure 10 illustrates the interrogation experiment set-up. Two ^4He detectors were positioned at 1.2 meters from the D-D neutron generator. Assuming the

Figure 9: The ratio between the maximum amplitude of the slow component (M_{slow}) and the fast component peak (M_{fast}) is calculated for the neutron and gamma events of ^{252}Cf respectively.

Figure 10: Experiment schematic of the D-D neutron generator active interrogation on natural uranium samples. The ^4He detectors measure both the interrogating neutrons and induced fission neutrons.

neutron source was isotropic and emitting 10^9 neutrons/second, an MCNPX-PoLiMi [14] simulation of the experiment was done. The detector was modeled with pure ^4He gas without other materials such as the wavelength shifter. About 4.27×10^6 neutrons were transported into the two detectors and 40882 neutrons had elastic scattering interactions with the ^4He inside the detector. Next, three natural uranium samples were put in front of the ^4He detectors and subjected to the interrogating D-D neutrons of 2.45 MeV. The uranium samples are encased

in aluminum cylinders (2.85 cm in diameter, 21 cm in length) and each weighs 2 kg. Using MCNPX-PoLiMi and 10^9 source neutrons, the ratio of $^{238}\text{U}(\text{n},\text{f})$ and $^{235}\text{U}(\text{n},\text{f})$ reaction rates in the natural uranium samples was found to be 18:1. Among the 40152 neutrons which underwent elastic scattering with ^4He , about 1.9% (i.e. 750 neutrons) originated from fission reactions. Finally, the simulation showed only 21 fission neutrons (or 0.052%) that deposited energies greater than 1.57 MeV, the maximum deposition energy of D-D neutrons.

During the actual operation, the D-D neutron generator at the University of Florida generated between 10^7 and 10^9 neutrons per second. At approximately the same distance as the ^4He detectors from the generator, a neutron dose of 700 mRem/hr and a gamma dose of 60 mR/hr were measured with a ^3He neutron survey meter and a Geiger-Muller counter, respectively. The two ^4He detectors recorded 2.56 million events in 6 minutes. Pile-up and spurious trigger rejection algorithms were applied and Figure 11 shows the density plots of the filtered results. If unfiltered, pile-up events would contribute to a higher count of neutron events, which are identified by the large integrated slow component values. The other filtered events include those triggered by random PMT coincidences and having unreliable baseline pulses. The remaining good events (about 72% of total events) obtained by the filter algorithms show excellent gamma/neutron discrimination.

Figure 11: Density plots of events recorded by ^4He detectors during neutron generator operation. Pile-up and other (i.e. spurious trigger and bad baseline) filters were applied.

200 Compared to the generator signal, the fission neutron signal when natural
 uranium samples were interrogated by D-D neutrons was much smaller due to
 the small amount of fissionable materials. As the dominating signals were still
 from the generator and background sources, the same filter algorithms were ap-
 plied to the measured data. Figure 12 shows the density plots of the filtered
 205 results when the natural uranium samples were present. The total measurement
 time was 32 minutes with 12.5 million events, compared to the 6-minute mea-
 surement time for the recording of generator-only signal. The percentages of
 pile-up and other rejected events were 11% and 14% respectively. The density
 distributions in Figure 12 are similar to those in Figure 11 since the recorded
 events were dominantly the generator signal.

Figure 12: Density plots of events recorded by ^4He detectors during the active interrogation of natural uranium samples with a neutron generator. Pile-up and other (i.e. spurious trigger and bad baseline) filters were applied.

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3.1. Pulse height spectra

Pulse height spectrum is obtained by summing the fast and slow components of the ^4He detector outputs. For the generator neutrons, the pulse height spectrum is affected by pile-up and spurious trigger events due to the high back-
 215 ground radiation associated with the generator operation. Shown in Figure 13, the generator signal's pulse height spectrum would have been skewed to much higher values without any pulse filtering. Similarly, the pulse height spectrum

of the combined generator and induced uranium fission neutron signal has to be filtered as shown in Figure 14. It can be seen that pile-up events reach over
220 to the region of large pulse heights where the real higher energy neutron events are detected.
are detected.

Figure 13: Pile-up and other (for spurious trigger and bad baseline) filters are applied to the events recorded by ^4He detectors during generator operation. Pulse height spectra of corresponding events are plotted.

In order to verify the contribution of fission neutrons in the pulse height spectrum shown in Figure 14, a $125\ \mu\text{Ci}$ ^{252}Cf spontaneous neutron source was measured with ^4He detectors using the same calibration settings as in the active
225 uranium interrogation case. A total of 0.12 million events were recorded in 5 minutes. Pile-up and spurious trigger filters were applied to filter out 8% of the total events to prevent inaccurate pulse height calculations. It was observed that such events in general had low pulse heights and hence did not affect the determination of the maximum pulse height spectrum value as the bounding
230 energy deposition value for ^{252}Cf neutrons. Finally, the filtered pulse height spectrum from Figure 14 is plotted against those of the filtered generator-only signal and ^{252}Cf in Figure 15. The good agreement between ^{252}Cf and high-

Figure 14: Pile-up and other (for spurious trigger and bad baseline) filters are applied to the events recorded by ^4He detectors during active interrogation of natural uranium by generator neutrons. Pulse height spectra of corresponding events are plotted.

energy fission neutron pulse heights verifies the energy of the fission neutrons unambiguously.

235 As a result of the ^4He detector re-calibration and post-processing of events, the efficiency of fission neutron detection is greatly improved. In Figure 15, there are 1359 neutrons (background subtracted) whose pulse height values are above 3.26×10^4 , the cut-off value set by the generator-only pulse height spectrum. The efficiency of ^4He detectors to discriminate fission neutrons is calculated
240 as the ratio of 1359 and about 8.79×10^6 , the combined number of generator and fission neutrons after applying a simple linear pulse shape discrimination (PSD). A more conservative PSD was also applied to remove the possible gamma counts as false neutrons, as shown in Figure 16, which resulted in a smaller neutron count of about 8.54×10^6 . Table 1 shows the neutron counts and the
245 corresponding efficiencies for the low and high gain settings, with and without event filters. There were fewer events in the high gain case because of a shorter measurement time (11 minutes versus 32 minutes in the low gain case) and the

Figure 15: ^{252}Cf spontaneous fission neutrons and the induced uranium fission neutrons ('Generator + U') have the same range of pulse heights above the cut-off value set by the 'Generator' spectrum. Counts above the cut-off are treated as the background unrelated to the DD generator and hence will be subtracted from the 'Generator+U' counts for the calculation of induced fission neutron counts.

neutron generator flux level was not uniform throughout all measurements. In both high and low PMT gain settings, the event filters were able to improve the efficiency of differentiating fission neutrons. At the higher gain, the saturation of fast component scintillation in the DAQ was a concern for the calculation of pulse height values of high energy events such as induced fission neutrons. For the sole purpose of event counting (but not for source spectrum unfolding) in this work, saturated pulses were not filtered as the other unreliable events and 36% of the 341 counts above the pulse height cut-off value set by the generator signal has saturated fast components. At the low gain setting, only 3% of the 1359 counts were saturated pulses. A combination of lower PMT gain, higher trigger threshold and filter algorithms result in the preferential triggering of events with high deposition energies by fission neutrons and offers the highest efficiency.

Figure 16: Pulse shape discrimination of the measured events by ^4He detectors can be done by a linear or conservative cut which excludes indistinguishable events of either gammas or neutrons.

Table 1: Efficiency is calculated as N_f/N_{total} where N_f is the count of neutrons above the pulse height cut-off line with background count subtracted, and N_{total} is calculated depending on a simple linear PSD cut or a more conservative cut shown in Figure 16. At lower gain (LG) value and with pulse filtering (w/ F), N_f separable from the generator neutron signal is the largest. LG and high gain (HG) N_{total} were taken with different measurement durations.

	N_f	$N_{total} (\times 10^6)$	Efficiency (%)
LG w/ F	1359	8.54 $\sim$ 8.79	0.0155 $\sim$ 0.0159
LG w/o F	602	11.24 $\sim$ 11.57	0.0052 $\sim$ 0.0054
HG w/ F	341	4.37 $\sim$ 4.44	0.0077 $\sim$ 0.0078
HG w/o F	214	6.01 $\sim$ 6.12	0.0035 $\sim$ 0.0036

The measured efficiency of 0.0155 $\sim$ 0.0159 % is lower than the PoLiMi-calculated efficiency of 0.052%. The greater value of the latter is due to the over-simplification of both the ^4He detector and generator source models. Structural components such as the stainless steel casing of the detectors were not modeled and there was the inexact knowledge of the generator output level,

the associated gamma/X-ray production and the solid angle subtended by the detectors. In addition, fission neutron events which were embedded in spurious trigger and pile-up events (such as in Figures 5 and 7) were inadvertently filtered out as well. While it is difficult to make an absolute comparison between the simulation and measurement results, the improvement of efficiency achieved by experimental calibration and post-processing filters is more relevant for the further development of an active interrogation system based on the ^4He detectors. A different pulse height cut-off value for differentiating induced fission neutrons can also be examined with other interrogating sources such as a Deuterium-Tritium neutron generator or lower energy (<1 MeV) neutrons. They have been used for active interrogation measurements in conjunction with liquid scintillators [15, 16] which are also capable of PSD and neutron spectrometry. However, the light yield of gamma events in liquid scintillators can be ten times higher for the same energy deposition than in ^4He detectors [8], such that the removal of pile-up, the amount of gamma shielding and n/ γ pulse shape discrimination are going to be important factors for the performance comparison between ^4He and liquid scintillators.

4. Conclusion

This work presents the measurement and pulse analysis techniques for the ^4He fast neutron detectors in order to measure a weak fission signal amid a strong neutron and gamma background. Higher energy fission neutrons cause more scintillation light in ^4He than the 2.45 MeV D-D interrogating neutrons. The difference in the scintillation pulse height spectra is used to discriminate interrogating neutrons from the induced uranium fission neutrons. The calibration of the ^4He detectors in terms of lowering the PMT gain and increasing trigger threshold has been done to increase the ^4He detector system's sensitivity to fission neutrons whose energies are greater than the interrogating neutrons. Pile-up and other unreliable events associated with the high count-rate interrogating background have been filtered out in the post-processing of detector

295 outputs. Both the detector efficiency and the accuracy of the pulse height spec-
trum in the energy range above 2.45 MeV are improved. The measured pulse
height spectrum of induced uranium fission neutrons matches that of ^{252}Cf
spontaneous fission neutrons, demonstrating the ^4He detector's capability of
discriminating fast neutron energy above the energy of D-D neutrons and up to
300 10 MeV.

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