NSDD
NUCLEAR SMUGGLING DETECTION AND DETERRENCE

Investigating Gadolinium for Neutron Detections using Gamma-Ray Detectors in Mobile Applications

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1. Introduction

1.1 Motivations

For enhancing the effectiveness of nonproliferation efforts in neutron detection, most portable instruments rely on ^{6}Li scintillators, ^{10}B-based detectors, or gas-filled ^{3}He proportional counters. Additionally, gamma-ray detectors based on scintillators and semiconductors are often employed for search applications to find radioactive material in the field. These systems typically include dedicated detectors along with separate high voltage supplies and processing electronics for the gamma-ray and neutron detectors. Ideally, a portable radiation detection system should be lightweight, compact, and cost-effective.

In the field, scintillators can serve a dual purpose: (1) detecting gamma-rays and (2) detecting neutrons. Gamma-ray detection with scintillators is based on the interaction of gamma-rays within the scintillating material, whereas neutron detection depends indirectly on neutron capture events. These capture events generate conversion electrons and gamma-rays, which can interact with the scintillator. For enhancing neutron capture, the scintillator can be surrounded by neutron absorber materials with a high neutron cross section. The resulting secondary electrons and gamma-rays from neutron interactions, depending on the absorber material used, can then be analyzed to detect the presence of neutron sources. Similarly, semiconductor-based detectors can be employed along with neutron absorbers as liners for neutron detection.

^{158}Gd has a significantly larger neutron cross section than ^{3}He, commonly used in gas-filled proportional counters, as shown in Figure 1 [1]. For thermal (0.025 eV) neutrons, the absorption cross section of ^{158}Gd is 10,000 times greater than that of ^{3}He (refer to Figure 1). This feature makes naturally occurring gadolinium, which consists of 24.8% ^{158}Gd, a promising neutron absorber material for use in combination with gamma-ray detectors–yielding a hybrid detector–for neutron detection.

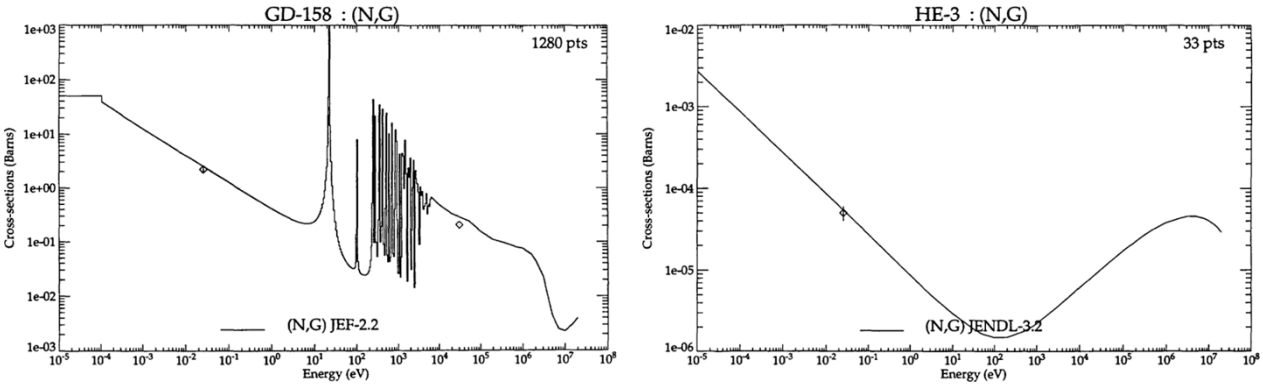

Figure 1. Neutron capture cross sections of ^{158}Gd and ^{3}He as a function of the neutron energy.

1.2 Goals

This report presents the analysis on evaluating the potential of natural gadolinium as a neutron absorber for neutron detection in the field. The study focused on using sodium iodide (NaI) and high-purity germanium (HPGe) detectors to measure neutron sources under three different configurations: (1) a bare neutron source, (2) a neutron source with gadolinium, and (3) neutron source with gadolinium and polyethylene placed in between. For the second and third configurations, a gadolinium sheet measuring 100 mm x 100 mm x 2 mm was used, with the 2 mm thickness positioned directly in front of the

detector. Moreover, for the third configuration, polyethylene with a thickness of 0.0508 m (2”) was used to surround the neutron source. The neutron source and detector

2. Procedure

This section outlines the steps undertaken in this study to evaluate gadolinium’s effectiveness in neutron detection.

2.1 Gadolinium Characterization

The gadolinium sheet obtained for the study was characterized for quality assurance using a scanning electron microscope (Figure 2). The analysis showed that the sheet was primarily composed of gadolinium (97%), with minor contaminants of carbon (2%) and oxygen (1%).

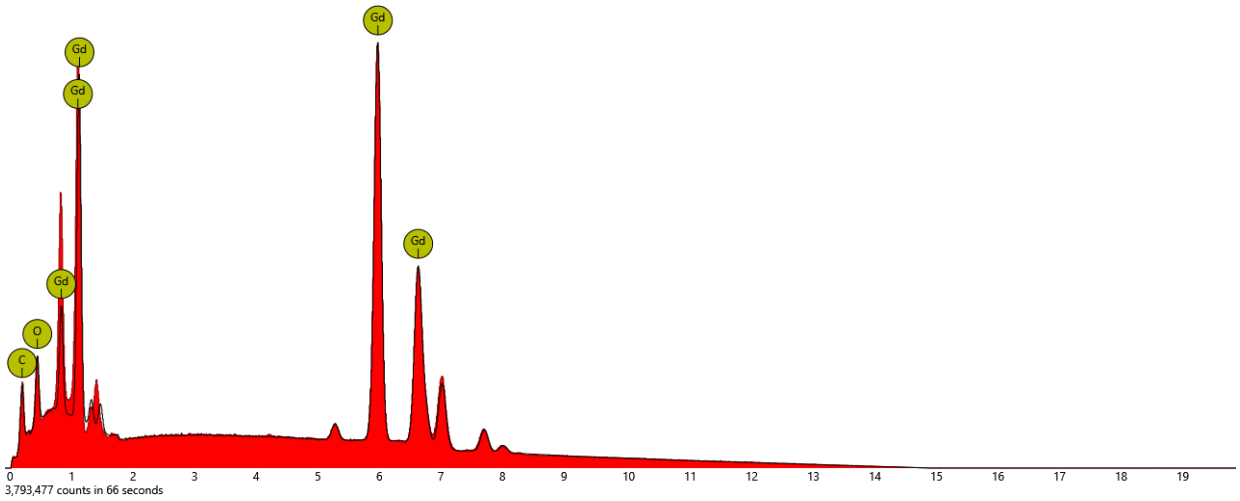

Figure 2. Analysis of gadolinium sheet using scanning electron microscope.

2.2 Setup

Figure 3 shows a top view of the measurement setup for the third configuration, where the neutron source is placed 1 m away from the detector face. A laboratory-based ^{252}Cf source, emitting approximately 20,000 neutrons per second, was used as the neutron source. The source was surrounded by polyethylene, while the gadolinium sheet was placed directly in front of the detector face. For covering the face the NaI detector, four gadolinium sheets were used – offering a total surface area of 0.04 m^2 for neutrons, whereas three sheets with a total surface area of 0.03 m^2 were used for the HPGe detector.

In the NaI detector setup, the NaI crystal was connected to a photomultiplier tube, which was then coupled to a digiBASE multichannel analyzer. This system was routed to a computer running the MAESTRO software program for collecting data and analyzing the gamma-ray spectrum. The spectrum, which represents the detector’s response, was calibrated using a ^{152}Eu radioactive source.

Similarly, in the HPGe detector setup, an ORTEC trans-SPEC-DX-100T HPGe spectrometer was connected to the computer via USB, allowing the MAESTRO software to save the collected spectra. A test spectrum with ^{235}U and ^{137}Cs sources confirmed the auto-calibration was done correctly, not requiring manual calibration.

The gamma-ray spectrum was collected using both detectors over a measurement time of five minutes.

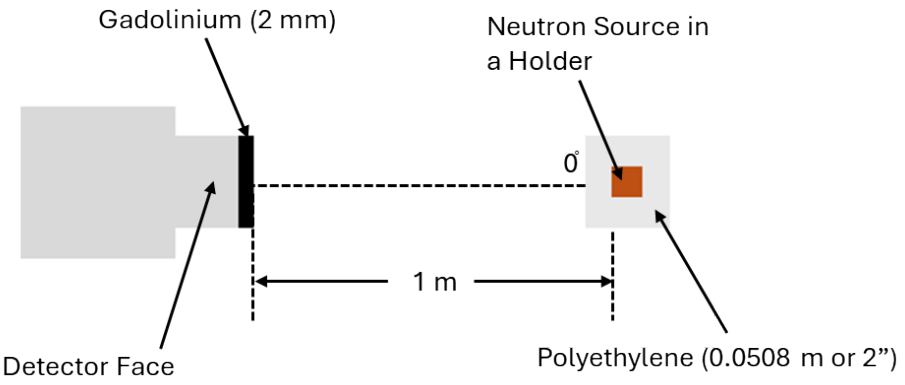

Figure 3. Top view of the measurement setup (not to scale) for the third configuration, with polyethylene encompassing the neutron source and gadolinium sheet in front of the detector face.

2.3 Gamma-Ray Peaks

As neutron capture by gadolinium produces cascades of secondary electrons and gamma-rays, a set of representative gamma-ray peaks was identified. For the analysis of the collected spectra, the following gamma-ray peaks were used [2], [3]:

(1) 199 keV,
(2) 898 keV,
(3) 944 keV,
(4) 959 keV,
(5) 1010 keV,
(6) 1107 keV,
(7) 1187 keV,
(8) 1818 keV, and
(9) 2073 keV.

3. Results

This section presents the measurement results using each detector system across the three different configurations described earlier.

3.1 NaI

Figure 4 shows the gamma-ray spectrum obtained using the NaI detector for two configurations: (1) a bare neutron source and (2) a neutron source with gadolinium. The results for the third configuration, where polyethylene was placed between the neutron source and the gadolinium sheet, were similar to those of the second configuration. From these measurements, three key observations were:

- The gadolinium sheet was found to contain trace amount of ^{176}Lu, which emits 201.8 keV and 306.8 keV gamma-rays. This conclusion was drawn from the observed gamma-ray peaks around 200 keV and 310 keV (marked by pairs of red dotted lines in Figure 4).
- The spectrum (shown in black) displays a reduction in low-energy gamma-ray contributions. This reduction is attributed to the gadolinium sheet, where its 2 mm thickness blocks many of the low-energy gamma-rays.

- No distinct gamma-ray peak associated with neutron capture by gadolinium was observed in a five-minute measurement. The observed peaks in Figure 4 are due to either californium (the 388.2 keV peak is due to trace contamination of ^{249}Cf in the sample) or ^{176}Lu (201.8 keV and 306.8 keV peaks).

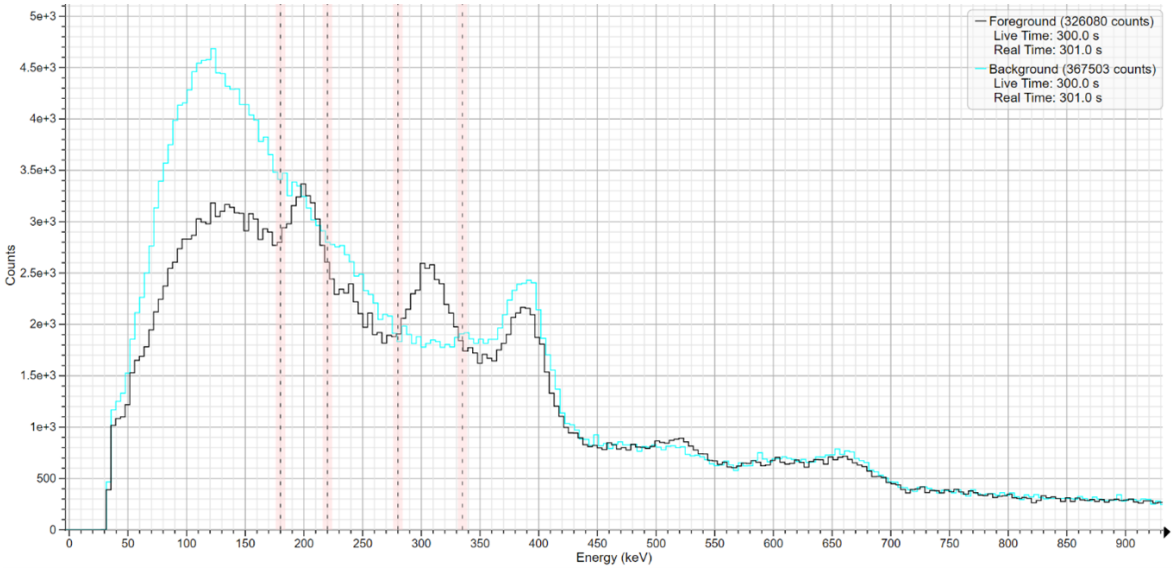

Figure 4. The gamma-ray spectra for a bare neutron source (shown in blue as background) and a neutron source with gadolinium (shown in black as foreground) using the NaI detector.

3.2 HPGe

Figure 5 shows the gamma-ray spectrum obtained from all three configurations using the HPGe detector. Similar to the results with the NaI detector, none of the nine gamma-ray peaks were evident in these measurements. This outcome was primarily attributed to three factors: (a) the intensity of neutron source, (b) the measurement time, and (c) the thickness of the gadolinium sheet, all of which contributed to very poor detection efficiency.

For a gamma-ray of 200 keV, using the gamma-ray attenuation formula $I = I_o * e^{(-\mu x)}$, the total mass attenuation coefficient (not accounting for coherent scattering) of 0.5123 cm^2/g, and the gadolinium density of 7.9 g/cm^3, the attenuation through the 2 mm thick gadolinium sheet is 55%. Thus, the gadolinium sheet blocks about half of all 199 keV gamma-ray generated from the neutron capture events. This attenuation phenomenon is much lower for the high energy gamma-rays, about 9% for the 1 MeV gamma-rays.

For investigating these factors, an additional set of measurements was performed in this proof-of-concept analysis. These measurements using the HPGe detector utilized a much stronger neutron source–emitting around 700,000 neutrons per second, approximately 37x higher than the original source–and extended the measurement time to 30 mins, six times longer than the initial setup. Figure 6 shows the gamma-ray spectrum for all three configurations under the new setup. In the second configuration, which represents a neutron source with a gadolinium sheet, six of the nine gamma-ray peaks were automatically identified by the *Peak Manager* in Interspec software. The remaining three peaks were manually added (with all nine peaks highlighted in blue, though some are not clearly visible). The net gamma-ray count across these peaks was measured at approximately 2 gamma-rays per second.

Out of the 700,000 neutrons emitted per second in all directions, the neutron incident rate at a point 1 m away is estimated to be 55,000 neutrons per second per square meter. Given that one gadolinium sheet has an area of 0.01 m^2, the neutron incident rate on three sheets is approximately 1,700 neutrons per second. By comparing the measured gamma-ray count rate (2 counts per second) to the incident neutron rate (1,700 incident neutrons per second), the detection efficiency is calculated to be less than 0.2%. This low efficiency makes a gadolinium-wrapped gamma-ray detector a poor neutron detector, especially in mobile applications.

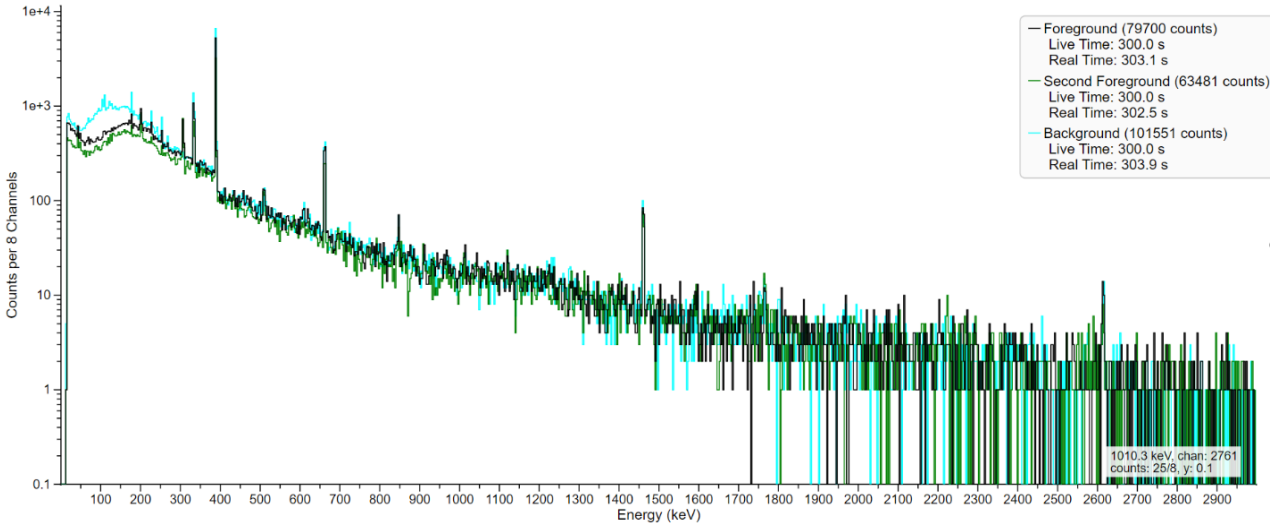

Figure 5. The gamma-ray spectra for a bare neutron source (shown in blue as background), a neutron source with gadolinium (shown in black as foreground), and a neutron source with gadolinium and polyethylene (shown in green as second foreground) using the HPGe detector.

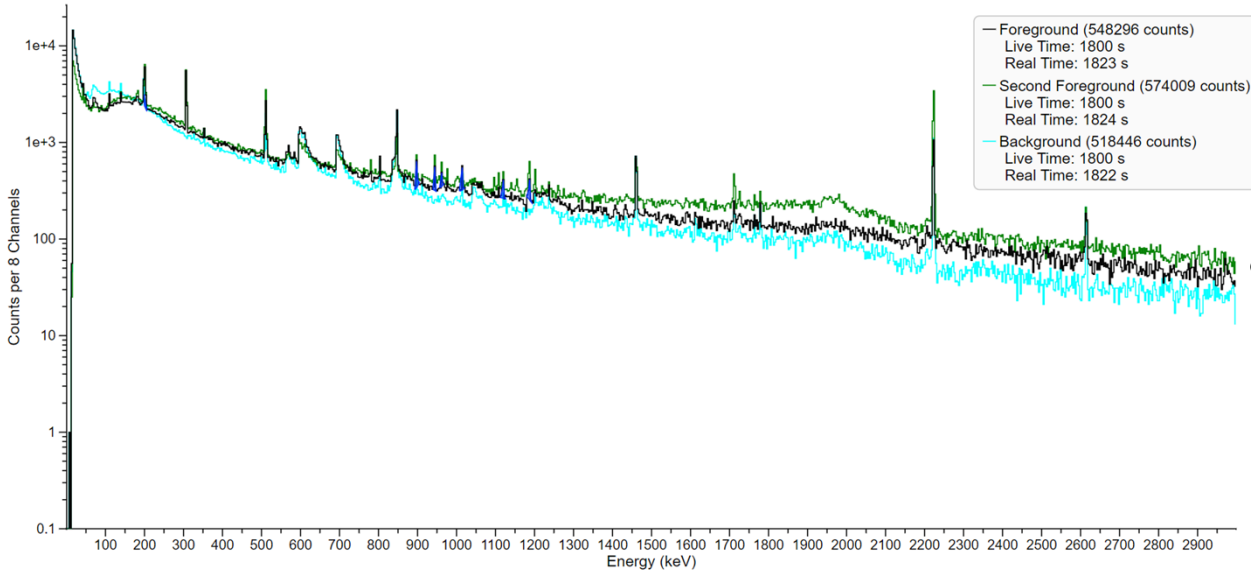

Figure 6. The gamma-ray spectra for all three configurations under the new setup using the HPGe detector (same color scheme as in Figure 5).

As seen in Figure 5 and Figure 6, the addition of polyethylene had little effect on increasing the neutron capture events in gadolinium and in turn, to the gamma-ray spectrum. The presence of the polyethylene

is most observable via the 2,223 keV peak due to neutron capture in hydrogen and the associated Compton edge features – thus demonstrating polyethylene to be a better neutron indicator for gamma-ray based detectors than gadolinium, despite gadolinium's enormous absorption cross section.

4. Conclusion

Gadolinium as a neutron absorber for field-based neutron detection was assessed using NaI and HPGe detectors in various configurations. In this study, a 2 mm thick gadolinium sheet with a total surface area of 0.03-0.04 m^2 was positioned directly in front of the detector face. A scanning electron microscope examination confirmed that the sheet's major constituent was in fact gadolinium (at least 97%).

This proof-of-concept analysis assessed three different configurations with each detector: (1) a bare neutron source, (2) a neutron source with gadolinium, and (3) neutron source with gadolinium and polyethylene placed in between. These measurements revealed trace amounts of ^{176}Lu in the gadolinium sheet, but no expected gamma-ray peaks from neutron capture events in gadolinium were evident. This lack of detection was primarily attributed to three factors: (a) the intensity of neutron source, (b) the measurement time, and (c) the thickness of the gadolinium sheet. While gadolinium has a high neutron absorption cross section, the emitted gamma-rays were either insufficient, secondary electrons were captured within the gadolinium sheet, or both–leading to very poor detection efficiency.

For investigating these factors, an additional set of measurements was performed using the HPGe detector. In these measurements, a stronger neutron source and longer measurement time were employed, which yielded more detectable results. The Interspec software successfully identified six of the nine expected gamma-ray peaks. However, the total gamma-ray count across these nine peaks was measured at only 2 counts per second, which is relatively low and indicates that longer measurement time would be necessary–something that may not always be feasible in field conditions. This measured count rate translates to a detector efficiency of less than 0.2%.

While the detector efficiency is low, it demonstrates a potential of using gadolinium in combination with gamma-ray detectors for neutron detection in mobile applications. Future analyses could focus on evaluating thinner gadolinium sheets, such as 0.25 mm, 0.5 mm, and 1 mm, to further characterize the effectiveness of gadolinium as neutron absorbers in hybrid detector systems. Polyethylene, with its 2,223 keV neutron capture gamma-rays, is also a viable alternative for using gamma-ray detectors to indicate the presence of a neutron source.

References

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