Microfabrication of a gadolinium-derived solid-state sensor for thermal neutrons

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ABSTRACT

Neutron sensing is critical in civilian and military applications. Conventional neutron sensors are limited by size, weight, cost, portability and helium supply. Here the microfabrication of gadolinium (Gd) conversion material–based heterojunction diodes for detecting thermal neutrons using electrical signals produced by internal conversion electrons (ICEs) is described. Films with negligible stress were produced at the tensile-compressive crossover point, enabling Gd coatings of any desired thickness by controlling the radiofrequency sputtering power and using the zero-point near p(Ar) of 50 mTorr at 100 W. Post-deposition Gd oxidation–induced spallation was eliminated by growing a residual stress-free 50 nm neodymium-doped aluminum cap layer atop Gd. The resultant coatings were stable for at least 6 years, demonstrating excellent stability and product shelf-life. Depositing Gd directly on the diode surface eliminated the air gap, leading to a 200-fold increase in electron capture efficiency and facilitating monolithic microfabrication. The conversion electron spectrum was dominated by ICEs with energies of 72, 132 and 174 keV. Results are reported for neutron reflection and moderation by polyethylene for enhanced sensitivity, and γ- and X-ray elimination for improved specificity. The optimal Gd thickness was 10.4 μm for a 300 μm-thick partially depleted diode of 300 mm² active surface area. Fast detection (within 10 min) at a neutron source-to-diode distance of 11.7 cm was achieved with this configuration. All ICE energies along with γ-ray and Kα0 X-rays were modeled to emphasize correlations between experiment and theory. Semi-conductor thermal neutron detectors offer advantages for field-sensing of radioactive neutron sources.

KEYWORDS: gadolinium converter, thermal neutron sensor, semi-conductor, microfabrication, device development, testing, solid-state neutron sensor

INTRODUCTION

Based upon their energies, neutrons are classified as fast (1–20 MeV, ~5 × 10⁶ m/s), slow (~1–10 eV, ~4 × 10⁴ m/s) and thermal (~0.025 eV, ~2 × 10³ m/s). Detection of a neutron is an indirect process based on its reactions with nuclei in other elements (‘conversion materials’), which generate energetic charged particles that can be detected by their electrical signal. For example, gadolinium (Gd) neutron capture reaction results in the emission of prompt gamma rays (γ-rays), internal conversion electrons (ICEs), Auger–Coster–Kronig (ACK) electrons and X-rays [1], all of which are detectable. Neutron sensors have civilian and military applications, and there is interest in robust, inexpensive neutron sensing. For example, personal or environmental monitoring can detect an increase in neutron levels due to an event, either accidental [2] or intentional [3]. Neutron-induced fission (‘neutron activation’) is used in nuclear reactors and nuclear weapons (NWs). 239Plutonium (239Pu) and 233,235Uranium (233,235U) are special nuclear materials (SNMs) and sources of spontaneous fission neutrons in NWs. Detection of SNMs is critical for nuclear non-proliferation and national security to track cargo at ports-of-entry. The low-energy
γ-rays of SNMs can be blocked, preventing detection. The α-particles with large linear energy transfer do not travel further than ~5.08 cm in air. Since neutrons penetrate γ-ray absorbers, an increase in the neutron level would be indicative of SNMs. Conventional neutron detectors use tritium (³H) decay product Helium-3 (³He), which has a reasonable cross-section for thermal neutrons. Besides there being a global shortage of ³H due to applications in national security, non-proliferation and medical diagnostics, ³He detectors suffer from issues of portability, high-bias voltage (1200–1800 V) and cost [4]. Semi-conductor devices (semi-conductor diodes, solid-state detectors) are potential replacements for ³He neutron detectors [5–7]. In this paper, the microfabrication of Gd converters on silicon (Si) diodes (for detecting thermal neutrons from the electrical signals of ICes) is described. Results are reported on neutron moderation, γ-ray elimination, sensitivity, specificity and speed of neutron detection. The data are modeled after earlier mathematical studies to emphasize correlations. A semi-conductor thermal neutron detector is suitable for field-sensing of neutron sources.

MATERIALS AND METHODS

Neutron source

The neutron source was a sealed container of the trivalent actinide ²⁵²Californium (²⁵²Cf), a neutron-emitting radioisotope (Serial # 1534, RS # 2852) from the radiological laboratory at Sandia National Laboratories (SNL). It is a high neutron emitter (3.768 neutrons/spontaneous fission, 3.1% decay probability and neutron yield of 2.314 × 10¹³ neutron/g/s), with a relatively long half-life of 2.645 years [8]. The majority (96.9%) of ²⁵²Cf undergoes α-decay, losing two protons and two neutrons, transforming into ²⁴⁸Curium (²⁴⁸Cm). Due to encapsulation, ⁴He nuclei do not escape the package. A small portion (3.092%) of ²⁵²Cf decays by spontaneous fission, producing neutrons, prompt γ-rays and photons [8]. The SNL ²⁵²Cf source had an activity of 38.4 μCi, within a cylindrical package 25 mm in length and 12.5 mm in diameter. The neutron sensor devices were characterized using this sealed neutron emitter.

Gd

Natural Gd used in this work was purchased from Plasmaterials (Livermore, CA) in the form of a 2" (5.08 cm) diameter, 0.125" (~0.32 cm) thick sputter target. The material was natural Gd with a purity of 99.9%. The rare earth metal was used as received.

Microfabrication

Oxidation-free, stable Gd films of uniform and precise thickness are critical for neutron sensing. Microfabrication enables such films to be created using rapid, consistent and large-volume manufacturing capabilities. Microfabrication was carried out in the cleanrooms of the Microsystems & Engineering Sciences Applications complex at SNL. Gd films were grown on 500-μm-thick Si(100) substrates or photodiodes using a radiofrequency (RF) sputter deposition technique [9]. Two deposition parameters were used to monitor and control film stress from tensile to compressive during sputter deposition: Ar sputter pressure and RF sputter power, which attenuate the kinetic energy of the depositing species while it migrates from target to substrate. Parenthetically, higher Ar pressure lowers the deposition rate, which is relevant when considering the deposition of films of up to 100 μm thickness. During sputtering, Gd and the electrodes were under vacuum and the inert gas argon (Ar) was introduced into the vacuum chamber as background. The RF power source was used to ionize Ar. The Gd target was bombarded by high-energy Ar ions, generating Gd ions, which then condensed on the substrate as a thin film of the desired thickness. An optimal area of uniform thickness was achieved by slowly rotating the substrate during the deposition process. Since the deposition rate is almost linearly proportional to RF power, Gd film deposition was carried out at 100 W power. Post-deposition Gd oxidation–induced spallation (peeling, flaking) was eliminated by growing a residual stress-free 50 nm neodymium (Nd)-doped aluminum (Al) cap layer atop the Gd film. All Gd depositions were performed in the absence of any type of adhesion layers on the substrate.

Surface profiling

Residual film stress was determined by measuring the change in curvature of a 5.08 cm Si(100) wafer using a profilometer before and after Gd deposition. An automated surface scanner, P-15 Profiler (KLA-Tencor, Milpitas, CA), which can profile a wide range of topographies, was used to scan the surface, using a capacitive measurement sensor in which the voltage is proportional to the surface height. The motorized stage enables automatic leveling and programmable rotation of the substrate, which is held by vacuum. The profiler was on a vibration-resistant table that suppressed low-frequency environmental noise. It is a sensitive surface profiler that measures step height, roughness and waviness on sample surfaces. The P-15 system uses diamond stylus-based scanning to achieve high resolution and can correlate submicron features with global surface measurements and image-processing techniques for noise reduction.

Neutron detection system

Testing and evaluation of the conversion of moderated neutrons into electrons by Gd was accomplished using the detector system illustrated in Fig. 1. It consisted of a ²⁵²Cf neutron source sitting atop a 5.1 cm polyethylene reflector. The neutron source was

![Fig. 1. Schematic of the experimental setup. This figure shows ²⁵²Cf source, HDPE moderator, steel, and Sn for γ/X-ray shielding, Gd film, and the PIN diode detector. The experimental setup is shown schematically in accordance with previous reports [5–7].](image-url)
progressively layered with 0.64 cm steel, 0.1 cm tin, a 3 cm polyethylene moderator and a 4 cm air-gap, before the placement of the 10 μm Gd-coated 500 μm-thick Si(100) wafer below the diode. This diagram illustrates the experimental set-up for uncoated diodes using the Gd on Si(100) wafer electron capture technique. For Gd-coated diodes, the wafer was removed and Gd was coated directly onto the diode [7].

**Partially depleted diodes**

Electrons produced by neutron capture of Gd were measured by Canberra Si diodes (Canberra Industries, Inc., Meriden, CT). These are partially depleted (PD), passivated, implanted, planar, silicon (PIPS) charged particle detectors with an implanted barrier contact that forms a thin abrupt junction. The diodes were in metal housings with entrance window of <50 nm for improved resolution, and 100–1000 μm depletion regions sufficient to stop 0.01–0.3 MeV ICEs. The rugged PIPS detectors were fabricated by planar processing using photolithographic techniques for defining device geometries. The PIPS detectors have a leakage current that is typically 1/8–1/10 of the Si surface barrier or diffused junction detectors and low reverse current, translating into low-noise. Size scaling was accomplished by various diode configurations; all devices had a 300 mm² active area, with 100–500 μm thicknesses and reverse-bias operating voltages of −40 V for 100 μm, −60 V for 300 μm and −100 V for 500 μm diodes. The nature of semi-conductor devices requires that they are sensitive to electrons as well as to extraneous γ- and x-ray radiation. Selective shielding of the detector using appropriate materials such as stainless steel can reduce the sensitivity of the device to electromagnetic ionizing radiation, but it cannot completely eliminate it. Thus, a differential operational mode will be demonstrated below that allows subtraction of the background while leaving the neutron’s effects intact.

**Electron nuclear spectroscopy**

Electron detection was accomplished using the electronic system illustrated in Fig. 2. The detection system consisted of amplification of the Canberra diode response to incoming conversion electrons from the Gd. The diode response was amplified using a Canberra 2003 BT preamplifier, which operates as a charge-to-voltage converter (0.45 V/pC; bias to ±1 kV). The current pulse was then Gaussian-shaped by the ORTEC 671 amplifier (ORTEC, Oak Ridge, TN) designed for use with Si-charged particle detectors to allow analysis of the energy by the pulse-shaping system, and the ORTEC 926 spectrometer fitted with an analog-to-digital converter board and Universal Serial Bus interface. This Multichannel Buffer equipment is a Nuclear Instrumentation Module designed for high-performance data acquisition in electron nuclear spectroscopy. The resulting spectrum was analyzed using ORTEC Maestro MultiChannel Analyzer Emulation Software. The final output is directly proportional to the collected charge.

**Electron measurements**

Measurements of the individual diodes and Gd films were made as follows. After assembly of the system without the Gd film, a 57Co source (8.88 × 10⁻⁵ μCi) and 241Am (9.68 μCi) source were inserted directly below the PIN diode for calibration. The ORTEC Maestro software was then configured to correctly identify the energy of the 59.9 keV γ-rays from the 241Am and the 122 kHz and 136 keV γ-rays from the 57Co source. Next, the 241Am and the 57Co sources were removed and the 252Cf source was placed as shown in Fig. 1, again without the Gd film. This produced a background measurement over the energy range of interest. As the detector was sensitive to γ-radiation as well as to electrons, it was necessary to subtract the background radiation. Thus, the measured electronic signal was directly proportional to the incident neutron flux. Next, various thicknesses of Gd films were used and the measurement was repeated. Data were collected and stored for several diode thicknesses and for several integration times. Additionally, a measurement was made with the same system including the Gd film with a 0.025 mm Al layer between the Gd film and the PIN diode. The Al film was grounded to shunt any electrons from the Gd to the detector as verification that the signal was due to electrons rather than from another γ source.

**Modeling and efficiency calculations**

Modeling of thermal neutron flux was carried out using the Monte Carlo code Geometry ANd Tracking (GEANT4) for the simulation of the passage of particles through matter. The software and source code are available from [http://geant4.org/geant4/index.shtml](http://geant4.org/geant4/index.shtml) (11 November 2016, date last accessed). The software simulation toolkit is for modeling both the detector and the physics of the passage of particles through matter [10]. It provides an object-oriented technology and follows an iterative incremental software process. Since GEANT4 is a toolkit, an extensive collection of data libraries and C++ classes exist. The user is expected to build their own application for running simulations in GEANT4. In modeling the neutron source, parameters were set up to get as close as possible to the experimental conditions. The number of particles in the experiment could be found by multiplying the neutron flux (vide supra) by the number of seconds within a 12-h measurement time. This resulted in more than four billion neutrons in the actual experiment. In order to save computation time, simulations were performed with only one billion neutrons, and they took ~6 days to execute. During modeling, an outer cylinder represented the
bounding universe and a smaller cylinder in the middle represented the experimental apparatus. The charged particle detector was modeled as a thin, 500 μm Si layer directly above the Gd layer [7]. The trajectories of protons, electrons and neutrons were color coded during modeling and a C++ code was used to define this geometry, along with other classes and header files used in GEANT4 application. Neutron flux was calculated as described in the next section and converted into the actual number of neutrons by multiplying by the specified measurement duration. Energy deposited on the charged particle detector was recorded and the data was visualized as a histogram.

RESULTS AND DISCUSSION

Neutron signal

The capture of thermal neutrons by Gd produces conversion electrons at several different energies that can be detected by a PIN diode. A PIN diode has a wide, undoped intrinsic semi-conductor region (larger depletion region, unlike PN diodes [7]) that is flanked by doped p- and n-type semi-conductors for ohmic contacts. The PIN diode enhances charge collection efficiency and response time of the planar conversion layer detector. The primary electron energies are approximately 72 keV, 132 keV and 174 keV. These electrons are ionized from the K and L shells of Gd atoms [11] as follows:

\[ ^{155}\text{Gd}(-15\%) + n \rightarrow ^{156}\text{Gd} + \gamma (0.09, 0.20 \text{ and } 0.30 \text{ keV}) + X-\text{ray} + \text{ACK} + \text{ICE} \]

\[ ^{157}\text{Gd}(-16\%) + n \rightarrow ^{158}\text{Gd} + \gamma (0.08, 0.18 \text{ and } 0.28 \text{ keV}) + X-\text{ray} + \text{ACK} + \text{ICE} \]

In 39% of neutron capture by Gd, ICEs with energy mainly of 72 keV (L shell) are emitted, and the conversion efficiency can reach up to 30% [12]. The \(^{252}\text{Cf}\) neutron source undergoes spontaneous \(\alpha\)-decay fission with the loss of two protons and two neutrons [8]. The neutron flux \(n\) for this source was calculated as, \(n = A \times (3.7 \times 10^{10} \text{ Bq/Ci}) \times (s. f.) \times \nu\), where \(A\) is the neutron activity in Ci (34.5 \times 10^{10} \text{ Bq/s}), s. f. is the branching ratio for spontaneous fission (0.03,092), and \(\nu\) is the average number of neutrons per fission (3.757). Using these values, a neutron flux of \(1.487 \times 10^5\) neutrons/s was calculated. This level of neutron activity was sufficient to generate measurements within an experimental time-frame.

Choice of Gd

The naturally occurring rare earth metal used as a neutron conversion material was Gd, which is composed of several stable isotopes: \(^{154}\text{Gd}, ^{155}\text{Gd}, ^{156}\text{Gd}, ^{157}\text{Gd}, ^{158}\text{Gd}, \text{ and }^{160}\text{Gd}\. \text{Of these, } ^{157}\text{Gd and } ^{155}\text{Gd have the largest thermal neutron capture cross-sections of any stable isotope: 255 000 barns and 65 000 barns respectively [12], which permit fast capture with smaller spatial separation, thereby reducing background. These two isotopes provide ~30% abundance in natural Gd (14.8% } ^{155}\text{Gd and 15.7% } ^{157}\text{Gd), with an average effective cross-section of 49 000 barns, compared with 5300 barns for } ^{7}\text{He. Previous simulation results showed that natural Gd possessed high neutron capture efficiency [5–7], even without resorting to } ^{157}\text{Gd-enriched material [6]. Gd is also best used as a metal instead of a scintillator compound such as gadolinium oxisulfate (Gd}_2\text{O}_2\text{S), since the metallic form is not as } \gamma\text{-sensitive and the signal from Gd}_2\text{O}_2\text{S is significantly lower relative to Gd metal [6, 7]. These data supported the use of natural Gd metal instead of the expensive } ^{157}\text{Gd [13].}

Microfabrication

The solid surface design used here provided a greater coverage area for Gd [14]. The deposition of several-microns-thick Gd films upon a substrate surface is not trivial since residual stress is a problem caused by the energetic deposition process. These stresses limit the critical thickness to <1 μm, above which films begin to delaminate. As Ar pressure increases, the kinetic energy of the depositing species decreases [15], resulting in lower residual tensile stress, until it crosses zero and turns compressive. Since the deposition rate is linearly proportional to the RF power, the deposition of Gd films at 100 W power was studied. Figure 3 shows the results for 500 nm-thick Gd films capped with 50 nm Nd-doped Al. A monotonic progression of the residual film stress from tensile to compressive with increasing Ar pressure during film deposition was observed. At high pressures (120 mTorr, Fig. 3), a flattening of stress was seen, indicative of Gd film buckling, and the films had a cloudy/milky appearance. Such undesirable effects were eliminated, and films with negligible stress were produced at the tensile-compressive crossover (zero) point, enabling coatings of any desired thickness by controlling the RF sputtering power and using the zero-point crossover.

![Fig. 3. Residual stress determination. This figure shows residual stress determination of 500 nm Gd films on Si(100) deposited at a constant 100 W RF sputter power as a function of Ar total pressure. All films were coated with a 50 nm layer of Nd-doped Al. A smooth curve fitting was used to profile the data using Kaleidagraph (Synergy Software, Reading, PA). Inset shows a photograph of the conversion (C) material with Nd-doped Al capping layer to eliminate post-deposition, oxidation-induced Gd degradation [32].]
near \( p(Ar) \) of 50 mTorr (Fig. 3) to produce uniform films. The films exhibited the highly polished mirror-like shiny appearance of a smooth surface. Profilometer measurements of curvature before and after Gd film deposition showed no changes, indicative of zero stress. Rare-earth metals such as Gd are known to oxidize readily. This was eliminated by an oxygen barrier of a Nd-doped Al capping layer. The films did not exhibit spallation (peeling, flaking) or delamination between 3 and 20 \( \mu \)m thickness, which is approximately three times thicker than the 7 \( \mu \)m Gd films obtained by etching with HCl [5]. Empirical data such as daily measurements of wafer curvature for several days following deposition, along with retention of the original smooth, shiny, polished, reflective, mirror-like appearance of the film, demonstrated that such coatings were stable for at least 5 years, with no signs of delamination. Microfabrication of low-stress, oxidation-free Gd films of any desired thickness, and excellent product shelf-life, were ideal for neutron sensing.

**Detector set-up**

The detector set-up in accordance with previous reports [5–7] is shown schematically in Fig. 1. There were several important factors regarding the detector set-up. First, \( \gamma \)-rays are ubiquitous in the background [14], arising from ambient X-rays caused by \( \gamma \)-radiation scattering, including Compton scattering [1]. This situation is exacerbated when compared with low natural neutron background fluence. There are two sources contributing to \( \gamma \)-ray background: external \( \gamma \)-rays accompanying \( ^{252} \)Cf and internal prompt \( \gamma \)-rays [16] (Eqs 1 and 2) due to Gd’s \(( Z = 64 \) high probability of interaction that produces moderate energy electrons by Compton, photoelectric and electron–positron pair processes [17]. These factors make it easy to record false positives when detecting SNMs, since an electron from \( \gamma \)-interaction is not easily distinguished from ICEs. This problem was overcome by using thin \((100–500 \mu \)m \)Si wafers, which have low \( \gamma \)-ray sensitivity [18]. The \( \gamma \)-ray rejection (\( \gamma \)-blindness) was further ensured by a steel layer between the neutron source and the diode (Fig. 1).

Next, the \( \sim \)200 keV \( \gamma \)-photons produced by \( ^{252} \)Cf \([ \text{at the rate of } 1.3 \times 10^{13} \text{ photons/(s-g)} ] \) were filtered by the steel plate. Since all neutrons are born at fast energies \(( \text{MeV} \) ), significant moderation was required to achieve a high probability of capture reactions. The most probable neutron energy of \( ^{252} \)Cf is 0.7 MeV, and the average energy is 2.1 MeV [8]. The conversion cross-section of natural Gd to such high-energy neutrons is about a million-fold lower compared with thermal neutron energy \((0.026 \text{ eV} \) ). Therefore, a moderator was employed to slow the high-energy neutrons by using the hydrogen-rich polymer, high-density polyethylene (HDPE) layer between \( ^{252} \)Cf and the detector (Fig. 1) [14].

The \( \sim \)2 MeV neutrons produced by \( ^{252} \)Cf \([ 4.4 \times 10^8 \text{ neutrons/(s-Ci) } ] \) for the 38.4 \( \mu \)Ci cylindrical source equaled \( \sim 1.7 \times 10^6 \text{ neutrons/s } \) total. However, since the source dimension was a \( \sim 12.5 \) mm cylinder, the total neutron flux that intersected the detector was significantly less. A planar geometry limits the reaction products reaching the detector, since charged particles are emitted in all directions [17]. In order to improve the system efficiency, a HDPE layer was placed beneath the source to reflect neutrons back towards the detector (Fig. 1). A tin (Sn) layer was placed above the steel to filter additional X-rays that were formed during \( \gamma \)-filtering in the steel. The moderated \( \sim \)0.1 eV neutrons [19] travelled through an air layer before finally reaching the Gd-coated Si wafer. The total source-to-detector distance was 11.7 cm, close to the 10.6 cm reported previously [14]. Since the distance between the Gd converter and the Si detector is critical in determining efficiency, and an air-gap causes sensitivity losses, direct deposition of Gd film on the diode surface contributed to an overall 200-fold improvement in efficiency [20].

**Gd emission spectrum**

A spectrum recorded from the charged particle detector is shown in Fig. 4A, with peaks at various energy levels due to electrons and X-rays. Clearly visible in the profile are the \( K_{\alpha} \)X-ray at 43 keV, along with three ICEs at \( \sim 72, 131 \) and 174 keV over collection times of 1–10 h. Under these conditions, a signal/background (S/B) of 40 was obtained (Fig. 4B). The Fig. 4A profile was calculated by first finding the difference in the running average of the signal (\( \mu_{\text{fl}} \)) to baseline (\( \mu_b \)) for a series \((m-n) \) of data records and then finding the average counts at that energy over the period of averaging. The data were then divided as a function of energy to arrive at the counts/energy data. The mathematical algorithm was:

\[
\frac{C}{E_n} = \left[ \frac{\sum_{n=1}^{N} \left( \sum_{j=m}^{n} \mu_{C_j}(E_n) - \sum_{j=m}^{n} \mu_{B_j}(E_n) \right) N}{E_n} \right],
\]

where the ratio of counts/energy in any ‘bin’ \( n \) is given in Eqn 3. By measuring the spectrum, even low neutron fluxes from solid-state devices could be detected in this architecture, which was capable of miniaturization for field-monitoring applications.

In order to obtain a clean electron spectrum, ICEs were blocked using Al foil, enabling background subtraction to yield the electron foreground (Fig. 5A), and study of cross-sensitivity, where Al was blocking electrons below 100 keV (Fig. 5B) [21]. The majority of ICEs had an energy of \( \sim \)72 keV from the \( L \)-shell of \( ^{158} \)Gd, and these were the principal electron–hole pair producers in the semiconductor. This broad peak was due to the electrons losing some energy during transit through the Gd film. Another option is a window discriminator \((40–100 \text{ keV} \) ) to electronically filter electrons, such as those produced by \( \gamma \)-rays, with energies outside this range [18]. Other electron peaks were at 131 keV and 174 keV energies, from \( K \) and \( L \) shell electrons de-excitation of 181.9 keV nuclear level of \( ^{158} \)Gd (Fig. 5A). Missing from this spectrum are 29 keV ICEs from the de-excitation of 158Gd, which is attributed to low-abundance relative to \( ^{158} \)Gd ICEs. This could be due to the 25–30 keV setting of the lower-level discriminator to reduce noise. The 29 keV ICEs would lose energy as they travelled through the foil and not be detected. There were also no ICEs from the de-excitation of \( ^{158} \)Gd, which is attributed to low-abundance relative to \( ^{158} \)Gd ICEs. Longer collection times might reveal these ICEs. Bias scaling was performed using a 100 \( \mu \)m-thick diode with calibration at each bias for optimization, and the data yielded adequate S/B of 4.0 at the 30 min collection point, with high specificity at low flux (Fig. 5C). Reverse biasing of the detector by an external applied voltage improves charge collection by increasing the
Clearly collection times were long; therefore, the focus shifted towards optimizing the Gd film thickness to improve neutron capture and maximize ICE generation.

Optimizing Gd thickness

The Si semi-conductor has a cross-section of ~2.24 barns for thermal neutrons and an effective mean free path of ~8.6 cm, making neutron detection using Si alone impossible. For this reason, a Gd converter layer was used (Fig. 1). In the planar diode configuration (Fig. 1), due to its high cross-section, a thin layer of Gd was used to create charged particles that escaped this layer and were detected as electronic pulses. If the conversion layer was too thick, ICEs might not have sufficient energy to escape and reach the detector volume for electron–hole pair generation. Too-thin Gd layers might not capture thermal neutrons efficiently. The goal was a Gd conversion layer thickness that maximized neutron capture, while having the highest ICE escape potential. The optimum Gd thickness was calculated based on the reaction cross-section, Gd density and the range of ICEs. Due to Gd’s large cross-section, a 50 μm film will be completely opaque to thermal neutrons; however, none of the ICEs will have sufficient escape energy. The cross-section of 255 000 barns for $^{157}$Gd translates into a mean free path of 1.3 μm for thermal neutrons. Thus, a 60 μm Gd layer will stop ~99% of neutrons. With natural Gd, a 5 μm film will allow ICEs to escape, but might not result in maximum neutron capture. The mean free path of thermal neutrons in natural Gd is ~7 μm, whereas the range
of 72 keV ICEs is >20 μm. Therefore, a 7–10 μm-thick Gd layer should absorb ~80% of incident thermal neutrons, and ICEs will still have sufficient escape energy [7]. Optimum conversion layer thickness was determined experimentally by correlating the ICE count rate as a function of Gd foil thickness (Fig. 6). Results confirmed that the maximum count rate was indeed between 7 and 10 μm. Film thicknesses flanking these values had lower count rates, indicative of either low neutron capture (3.3 μm) or fewer ICEs having sufficient escape energy (15.3 μm and 20 μm) (Fig. 6A). This decline is due to most of the neutrons being stopped in the lower depths of the Gd film, and the odds are against their escape as the film thickness increases [5]. The experimental data agreed with the theoretical calculations (below).

Using a mean free path of 6.7 μm for thermal neutrons, the probability of interaction, \( p \), is given by equation 4:

\[
p(x) = \int_0^x \sum_x e^{-x} \sum_x' dx'
\]

where \( x \) is the thickness of the material traversed by the neutrons. When Eqn 4 was profiled as a function of natural Gd thickness, the probability of a neutron absorption with 1, 2, 3 and 4 mean free paths could be determined (Fig. 6B). Results showed that ~80% probability of the interaction calculated theoretically (Fig. 6B) was at 10 μm Gd, the same as determined empirically (Fig. 6A). Although the probability of interaction increased slightly at >10 μm Gd (Fig. 6B), the escape potential of ICEs decreased simultaneously (Fig. 6A). All further experiments were therefore carried out using a 10 μm Gd layer, which was close to the 12 μm predicted previously for high efficiency [16].

Fig. 6. Gd film thickness optimization. Panel A shows Gd film optimization determined experimentally using conversion electron count rate from neutron capture in natural Gd at several different film thicknesses. Panel B shows the probability of neutron capture in natural Gd as determined theoretically. Probability is shown explicitly at 1 (6.7 mm), 2 (13.4 mm), 3 (20.2 mm), and 4 (26.9 mm) mean free paths.

Fig. 7. Integration time. Panel A shows the profile of four replicates of a 30 min. integration time. The trace above the baseline is a running average of the data and demonstrates the ability to clearly resolve the 72 keV, 132 keV, and 174 keV electrons produced by the moderated electron interaction with the natural Gd film. Panel B shows four replicates of a 10 min. integration time. The remainder of the details are the same as in panel A. The diode used in both experiments was the same and shown on top of the panels. The running averages are offset for clarity by adding 30 counts to separate them from the raw data.
Diode thickness
A 300 µm-thick diode gave the best sensitivity for neutron detection (Fig. 7), in accordance with a previous report [7]. There were several considerations in determining the optimum depletion layer thickness. Si does not detect neutrons—only ionizing particles such as electrons. The range of 70 keV ICEs in Si is ~25 µm [5]. For maximum charge collection, the Si layer should have a thickness at least equal to ~25 µm. With thinner diodes there could be signal loss, since the ICE range will exceed the Si thickness, leading to the ICEs escaping from the diode’s sensitive volume [5, 6]. The band-gap in Si of 1.11 eV results in an average energy of 3.6 keV for electron–hole creation [20]. A minimum ionizing particle creates on average 24 500 electron equivalents in a 300 µm-thick diode [18], enabling the measurement of a charge of ≤5000 electrons from the low-energy ICEs [20]. The choice of a 300 µm diode was consistent with literature reports that large depletion layers are preferable during ICE measurements [23], and the 300 µm diode was sufficiently thick to stop all ICEs and to tolerate noise [6].

Integration time
Integration time improved dramatically with optimized Gd film detectors (Fig. 7). All three dominant ICE energies, 72 keV, 132 keV and 174 keV, were detected within 30 min (Fig. 7A) and subsequently at 10 min (Fig. 7B), although the signal intensity was weaker, but nevertheless clearly discriminated. The signal intensity tracked linearly with integration time, with a three-fold decrease in the counts for 72 keV ICEs at 10 min (Fig. 7B) compared with 30 min (Fig. 7A). This time-scale fits reasonably with the notion that a 6.1 m container could be scanned in 10 min. However, it is not within limits for port-of-entry detection of SNMs, where containers are driven at 6–8 km h⁻¹, passing through a radiation portal monitor equipped with passive γ-ray scanners in ~20 s [24]. The 10-min detection window (Fig. 7B) is within striking distance of the 2-min scan for ~12 m containers [25]. The current detection time could be improved by decreasing/eliminating the air-gap (Fig. 1), and implementing other optimization steps. Alternately, the current configuration could be used as a confirmatory test for SNMs after alerting for suspicious cargo. Another possibility is to use the current detector for random cargo screening that accounts for <0.5% of incoming containers [24], and where scanning speed is not critical.

Modeling and efficiency
Intrinsic detector efficiency is the quotient of the number of events measured by a detector divided by the number of events incident on that detector and is governed by a series of probabilities of certain events [26]. These include the probability that a neutron will be captured by Gd, that every captured neutron will result in an electron emission, the escape probability of an electron from Gd and, finally, the probability of the escaped electron reaching the detector volume to produce an electrical signal [20]. Experimental results were modeled with excellent correlation to ICE production, based upon certain assumptions such as neutron stopping was maximal with 10 µm Gd, the detector was a planar configuration, and the converter stopped all ICEs [6]. Due to the large neutron capture cross-sections of ¹⁵⁵Gd and ¹⁵⁷Gd isotopes in natural Gd, it was further assumed that other reactions had negligible effects [5]. A visualization of the simulated experiment geometry and the results using GEANT4 are shown in Fig. 8A and B, with clear evidence of ICEs at 70–72 keV, along with Kα and Kβ X-rays in the simulation spectra. Successful simulation was demonstrated from the overlap of the modeling spectrum (Fig. 8A and B) and the experimental data (Figs 4A and 5A). Figure 6A shows the modeled electron count rate/area/time. With a 10 µm film, the ICEs signal was expected to be ~5 electrons/(cm² s). Figure 5A showed that the total integrated charge measured by the detector during Al blocking was ~450 electrons. Since the system was configured to integrate charge over 14 400 s, an intrinsic efficiency of ε = 0.002 was calculated for a 300 mm² detector. The intrinsic efficiency was defined as the number of electrons counts/s (10.8/s) divided by the number of neutrons incident on the detector/s. For simplicity, a point ²⁵²Cf source at a distance of R = 15 cm from the detector was used in Eqn 5, where det and source refer to the rates at the detector and source respectively, and ADET = 300 mm²:

\[
\varepsilon_{\text{det}} = \frac{\varepsilon_{\text{sys}}}{A_{\text{det}}} = \frac{\varepsilon_{\text{source}}}{A_{\text{det}}}.
\]

This yielded an intrinsic efficiency of εsys = 0.07, which compares well to literature values for ³He detectors [27, 28]. However, the system geometry (Fig. 1) alludes to ~1 cm separation between the detector and the Gd film. A significant improvement in electron capture efficiency should be possible by directly contacting the film with the diode surface [7]. This was indeed observed in Fig. 7, where total integrated charge was measured and ~78 000 electrons were counted from a 10 µm detector, yielding ε = 0.4: a 200-fold enhancement for electron capture efficiency.

![Fig. 8. GEANT4 Modeling. Panel A shows a model of the experimental geometry using GEANT4. The outer cylinder represents the bounding universe. The experimental apparatus is the smaller cylinder in the middle. Neutron trajectories are shown by wide traces. Smaller traces indicate the trajectories of protons and electrons, respectively. The charged particle detector is modeled as a 500 µm layer of Si directly above the layer of natural Gd. Panel B shows the simulation results. Arrows point to the evidence of Kα (42, 43 keV) and Kβ X-rays (48, 50 keV) along with major ICE at 72 keV, 131 keV, and 174 keV, respectively.](image-url)
CONCLUSION

The solid-state detector described here offers advantages over competing technologies. For example, the $^{10}$B (10B) neutron capture cross-section is 3840 barns, similar to that of $^3$He and an order of magnitude lower than that of natural Gd, leading to ~2.3-fold higher energy deposition rate with the latter [1, 22, 29]. The 2 MeV $\alpha$-particle from the $^{10}$B reaction has a range of 3.4 $\mu$m and the $^7$Li nucleus has an even shorter range [18]. The mean free path for thermal neutrons in $^{10}$B is ~20 $\mu$m. These conflicting factors reduce efficiency [5, 20]. On the other hand, Gd-capture cross-section extends to ~200 MeV, relative to $^{10}$B neutron energy moderation of 25–30 MeV [1]. The mean free path of thermal neutrons in Gd is 6.785 $\mu$m, and the range of 72 keV ICES is >20 $\mu$m. Thus, Gd converter layers could be made very thin and still capture ~95% of thermal neutrons and ICES having sufficient escape energy [20]. Unlike $^{10}$B, $^6$Li or $^3$He neutron detectors, solid-state Gd sensors do not produce energetic heavy charged $\alpha$-particles that interfere with detector electronics/software, degrade long-term sensor performance, or pose safety hazards [7, 16, 29]. Our emphasis was on low-damage electronics rather than $\alpha$-particles, which is compatible with rad-hard Si photodiodes [5–7]. There are ample reports of Gd being radiation resistant, enabling the fabrication of radiation-resistant containers for transportation, safe long-term storage and disposal of enriched nuclear waste [30, 31], leading to the conclusion that a Si-Gd detector would be robust and would have a long lifespan. In addition to $^3$He shortage [7], tube detectors are bulky, suffering from configuration, portability, and field deployment issues. High-pressure $^3$He makes transport difficult, and high-voltage bias is hazardous for underwater operations. System stability is affected by microphonic sensitivity, and simply bumping the system could produce false positives. Solid-state neutron detectors can be mass-produced, are compact for portability and field monitoring, and can operate on a few volts without requiring high power [23]. Our planar semi-conductor detector is a straightforward configuration for fixed or mobile neutron monitoring due to advantages of size [5], sensitivity, weight, power consumption, transportability, robustness, technological maturity [7], and manufacturing cost, and conventional wafer-level device microfabrication is possible for high throughput [13].

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CONFLICT OF INTEREST

The authors are not aware of any conflict of interest in this work or with the publication of this paper in the Journal of Radiation Research.

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