Experimental Determination of Gamma-ray Discrimination in Pillar-structured Thermal Neutron Detectors under High Gamma-ray Flux

Qinghui Shao^a, Adam M. Conway^a, Lars F. Voss^a, Radoslav P. Radev^a, Rebecca J. Nikolic^{a,*}, Mushtaq A. Dar^b, and Chin L. Cheung^c

^aLawrence Livermore National Laboratory, 7000 East Ave., Livermore, CA 94550, USA

^bKing Saud University, Riyadh 11421 Kingdom of Saudi Arabia

^cDepartment of Chemistry, University of Nebraska-Lincoln, Lincoln, NE 68588, USA

*Corresponding author. E-mail address: nikolic1@llnl.gov

Abstract

In this paper, we demonstrate a detector that has a high neutron-to-gamma discrimination of 8.5×10^5 with a high thermal neutron detection efficiency of 39% when exposed to a high gamma-ray field of 10^9 photons/cm²s. The detector is based on a silicon pillar structure filled with a neutron converter material (¹⁰B) designed to have high thermal neutron detection efficiency. The pillar dimensions are 50 μm pillar height, 2 μm pillar diameter and 2 μm spacing between adjacent pillars.

Keywords

Thermal neutron detector, Gamma-ray discrimination, Silicon pillar, Boron

Helium-3 tubes are the most widely used technology for detecting neutrons due to their high neutron detection efficiency as well as high gamma-ray discrimination, but there are issues with stability, sensitivity to microphonics, and very recently a shortage of helium-3 [1]. In order to find alternatives, solid-state thermal neutron detectors have been investigated that utilize various architectures and material combinations [2-7]. Many solid-state thermal neutron detectors are more sensitive to gamma-rays compared to helium-3 tubes, which can be an issue because the most nuclear materials emit ten or more times as many gamma-rays as neutrons. For example, in

measurements of spent fuel, gamma-ray fluxes of 1000 R/h ($\sim 10^9$ photons/cm²s) or more are encountered, and the gamma-ray discrimination of the detector may dominate all other considerations [8]. The electronic pulses generated by gamma-rays can be readily rejected by setting a high low-level-discriminator (LLD). However, a high LLD setting will limit the registration of low-energy pulses created by neutron events. The gamma-ray discrimination and intrinsic neutron detection efficiency are therefore important figures-of-merit for evaluating the performance of a neutron detector. In this paper, we report the performance of our pillar structured thermal neutron detector's gamma-ray discrimination properties under a high gamma-ray flux environment (up to 10^9 photons/cm²s). This is more than 10,000 times higher than the typical gamma-ray flux used for determination of gamma-ray discrimination in semiconductor based thermal neutron detectors [4-6, 9-12]. This is important because high fluxes can be associated with pulse pile up effects, which can limit the utility of this class of detectors.

¹⁰B is selected as the neutron converter material because of its high thermal neutron cross section of 3840 barns, compared to 940 barns of ⁶Li, which is also a popular neutron converter in solid-state thermal neutron detectors [2-3]. The thermal neutron capture reaction by ¹⁰B can be described as follows [13]:

The reaction byproducts deposit their energy into the semiconductor and generate electron-hole pairs. The charge carriers are swept out by an internal or applied electric field and collected by the electrodes for the registration of a neutron event. The selected semiconductor material should have a relative low atomic number (Z) and density (ρ) for the purpose of low gamma-ray interaction [13]. Silicon is used in our pillar detectors because it has both a low Z of 14 and ρ of 2.33 g/cm³. Murphy *et al.* reported that Si provides better gamma-ray rejection compared with other semiconductor materials, *i.e.* C (diamond), ZnO, GaAs, and CdTe [14].

Our Si-¹⁰B detector design uses a three-dimensionally integrated approach. The geometrical constraints on the converter material thickness are decoupled from the limitation of the ion track length [4-7]. The ¹⁰B thickness is defined by the pillar height, chosen as 50 μm , which is three

times the mean free path of thermal neutrons in ^{10}B . The pillar pitch of $4\ \mu\text{m}$ ($2\ \mu\text{m}$ pillar diameter and $2\ \mu\text{m}$ pillar spacing) is designed to allow a high probability of interaction between the energetic ions and the semiconductor pillars, created by nuclear reaction of the neutron with ^{10}B [4]. We have reported a 48.5% of intrinsic thermal neutron detection efficiency using this design [7]. Figure 1 (a) shows a schematic of a pillar structured thermal neutron detector. A silicon wafer comprised of a $3\ \mu\text{m}$ p^+ layer and a $47\ \mu\text{m}$ intrinsic layer (n^-) epitaxially grown on an n^+ substrate was used for device fabrication. The pillar diameter and spacing were defined lithographically followed by deep reactive ion etching to form a $50\text{-}\mu\text{m}$ -tall pillar array. A conformal ^{10}B coating was deposited to fill the gaps in the pillar array by chemical vapor deposition [15]. A Plasma Quest electron cyclotron resonance etcher was used for boron etching on the front side to expose the highly conductive p^+ pillar tops as shown in Fig. 1 (b). This was followed by surface planarization using Honeywell AccufloTM 2027 spin-on-polymer and chemical-mechanical polishing. Finally, metal contacts were formed by sputtering Al/Cr/Au ($5000\text{\AA}/500\text{\AA}/7500\text{\AA}$) on both sides.

Fig. 1 (a) Schematic of a pillar structured solid-state thermal neutron detector with $2\ \mu\text{m}$ pillar diameter, $2\ \mu\text{m}$ pillar spacing and $50\ \mu\text{m}$ pillar height (the dimensions are not to scale) and (b) scanning electron microscopy image of boron filled pillar arrays (top view).

In the pillar detector shown in Fig. 1, neutron absorption only takes place in the neutron conversion material (^{10}B), which is defined by the pillar height. Gamma-ray interaction takes

place in the entire Si portion, although only those absorbed in the intrinsic region contribute to the signal. Previously we investigated the effect of the intrinsic layer below the pillars on gamma-ray discrimination in a structure with a 25 μm intrinsic layer [6]. We found that the lowest gamma-ray response occurs when the thickness of intrinsic region below the pillars is reduced to a minimum, which limits the gamma-silicon interaction volume in the active region. Therefore, it is important to select a starting structure where the intrinsic layer thickness is close to the designed pillar height. In this paper, the pillars of the device under test (DUT) were formed by etching completely through the intrinsic region to increase the gamma-ray discrimination.

The experimental setup is shown in the inset of Fig. 2. The DUT was a $2 \times 2 \text{ mm}^2$ detector composed of a 50- μm -tall pillar array. It was attached to the side of a 5 cm-thick high-density polyethylene (HDPE) moderation fixture, which houses a ^{252}Cf source of $2.5 \times 10^{-2} \mu\text{g}$. The ^{252}Cf source primarily emits fission neutrons and had an activity of 13.44 μCi based on NIST calibration. The HDPE fixture moderates the high energy neutrons to produce thermal neutrons ($\sim 0.025 \text{ eV}$). The thermal neutron flux on the detector surface was $48 \text{ n/cm}^2\text{s}$. This is obtained by measuring the neutron spectra of a calibrated pillar detector having the same geometry ($2 \times 2 \text{ mm}^2$) with the same experimental setup. The thermal neutron detection efficiency of the calibrated pillar detector was obtained using two techniques as discussed in reference [7]. The gamma-ray flux on the detector surface was controlled by varying the distance between the detector and a large 662 keV ^{137}Cs gamma-ray source with an activity of 218.2 Ci. Data acquisition was carried out using an Ortec 142C preamplifier, an Ortec 572 shaping amplifier with a shaping time of 0.5 μs , and an Ortec AMETEK EASY-MCA-2K multichannel analyzer. The measurement live time was set to 20 minutes. The energy calibration of the electronic readout was performed by using external calibrated capacitors [16, 17].

96

97 Fig. 2 Measured background spectrum (no source), neutron spectrum and gamma spectrum of a 2
 98 $\times 2 \text{ mm}^2$ 50- μm -tall pillar detector. ^{137}Cs and ^{252}Cf were used as the gamma-ray source and the
 99 neutron source, respectively. The inset shows the experimental setup.

100 In order to investigate the gamma-ray discrimination at different flux levels, 1.37×10^6 and 1.37
 101 $\times 10^9$ photons/ cm^2s were obtained by positioning the DUT away from the ^{137}Cs source at
 102 distance of 632 cm and 20 cm, respectively. Fig. 2 shows the measured neutron and gamma
 103 spectra of the DUT using the geometry shown in Fig. 1. The fractional standard deviation of the
 104 neutron counting measurement is 3.2% obtained by $\sqrt{N/N}$, where N is the total number of
 105 recorded neutron counts. The intrinsic thermal neutron detection efficiency ($\varepsilon_{\text{int } n}$) is defined by
 106 dividing the number of counted neutrons in the neutron spectra by the number of thermal
 107 neutrons incident on the DUT. The intrinsic gamma-neutron detection efficiency ($\varepsilon_{\text{int } \gamma n}$) is
 108 defined by the number of gamma-rays counted as neutrons divided by the number of gamma-
 109 rays incident on the detector when no neutron source is present. Finally, we obtained the neutron
 110 to gamma-ray discrimination (n/γ):

111
$$n/\gamma = \varepsilon_{\text{int } n} / \varepsilon_{\text{int } \gamma n} \quad (2)$$

112 The relationship between $\varepsilon_{\text{int } n}$, n/γ discrimination and the LLD settings is shown in Fig. 3. Within

the LLD range of 67 to 107 keV, the two n/γ discrimination curves are close and they increase from 8×10^4 to 8×10^5 (gamma-ray flux: 1.37×10^6 photons/cm²s) and 4.6×10^4 to 8.5×10^5 (gamma-ray flux: 1.37×10^9 photons/cm²s), while the thermal neutron detection efficiency only decreases slightly from 41.0% to 39.0%. The LLD settings are high enough to reject any false neutron counts from X-rays with appreciable probabilities emitted from ²⁵²Cf, therefore a specific shielding against X-rays wasn't required.

Fig. 3 Measured intrinsic thermal neutron detection efficiency without a gamma-ray source and n/γ discrimination as a function of LLD with gamma-ray fluxes of 10^6 and 10^9 photons/cm²s.

The gamma-ray insensitivity of neutron detectors in Radiation Portal Monitor systems for homeland security applications requires that gamma radiation at exposure rate of up to 10 mR/h shall not trigger the neutron alarm as defined in American National Standards Institute ANSI N 42.35-2006 [18]. The gamma-ray flux converted from 10 mR/h is on the order of 10^4 photons/cm²s [13], much lower than the maximum flux reported here. As a benchmark, we set a LLD of 95 keV to ensure $\epsilon_{int \gamma n} \leq 10^{-6}$ as specified in PNNL report [19]. The corresponding $\epsilon_{int n}$ is 39.4%, and n/γ discriminations are 5×10^5 (gamma-ray flux: 1.37×10^6 photons/cm²s) and 3.5×10^5 (gamma-ray flux: 1.37×10^9 photons/cm²s). The measured n/γ discriminations are consistent with MCNP simulation results which are on the order of 10^5 based on the same device

structure [4].

As a comparison, K. -C. Huang et al. reported an n/γ discrimination of 2.4×10^4 of a 2.5×2.5 mm² ¹⁰B-Si honeycomb structured thermal neutron detector. This is based on the obtained $\varepsilon_{int n}$ of 26.1% and gamma sensitivity (intrinsic gamma-neutron detection efficiency) of 1.1×10^{-5} exposed to a ⁶⁰Co gamma-ray source of 10 mR/h (10^4 photons/cm²s) [9, 10]. As their hexagonal hole depth (45 μ m) in the honeycomb structure is close to our pillar height (50 μ m), a higher n/γ discrimination of 5×10^5 of the pillar detector could mainly be attributed to a relatively smaller Si volume portion of 20% in the active region compared to 46% in a honeycomb structure designed with 2.8 μ m hole diameter and 1 μ m Si wall thickness [9-11]. In back-to-back stacking of 1 cm² ⁶LiF-Si detectors developed in Kansas State University, an n/γ discrimination of 2.2×10^4 with a LLD of 300 keV and 1.0×10^6 with a much higher LLD of 450 keV using a ¹³⁷Cs source with a flux of 8.6×10^4 photons/cm²s were reported [12]. Due to the relatively low thermal neutron cross section of ⁶LiF, the Si trenches were etched 250 μ m deep for sufficient neutron absorption, significantly increasing the amount of Si which is sensitive to gamma-rays. The pillar structure has the least Si portion compared with the hole or trench structures; therefore the gamma-ray discrimination with a low LLD setting is higher.

In order to investigate the effect of the high gamma-ray field on the neutron detection performance, we measured pulse height spectra of the DUT in the presence of both ²⁵²Cf and ¹³⁷Cs, as shown in Fig. 4. As the gamma-ray flux increases, there is more than one gamma-ray striking the detector within the resolving time of the system, therefore the electronic pulse amplitude is the sum of several separate events (pulse pile-up). As a result, a high energy pulse is registered into the system. To accommodate the high count rate, the shortest available shaping time of 0.5 μ s in Ortec 572 shaping amplifier was used. Applying a reverse bias voltage can possibly reduce charge collection time by creating a high electrical field in the Si pillars. However, it also causes a high noise level associated with leakage current which negatively affects the neutron detection performance. Therefore, the detector was not biased during the measurements.

The “gamma-ray absolute rejection ratio for neutrons” (GARRn) measures the detector response in the presence of both a large gamma-ray source and a ²⁵²Cf neutron source. The GARRn value

will be unity if the gamma-ray source has no impact [19]. In our experiments, the GARRn value is extracted by the number of total neutron counts in the presence of both sources, divided by the number of total neutron counts in the presence of ^{252}Cf source only. To reject the pile-up pulses and meet the requirement of $0.9 \leq \text{GARRn} \leq 1.1$, the LLDs are set to 95 keV, 105 keV, 134 keV and 182 keV for the gamma-ray flux of 1.37×10^6 , 1.37×10^7 , 1.37×10^8 , and 1.37×10^9 photons/cm²s, respectively. Since the LLDs are set higher than 62 keV which is for ^{252}Cf only, some neutron counts at low energy channels are lost.

Fig. 4 Pulse height spectra of a $2 \times 2 \text{ mm}^2$ 50- μm -tall pillar detector exposed to a ^{252}Cf source and a ^{137}Cs source with varied fluxes. The gamma-ray fluxes are shown in parentheses.

To measure the lost portion of neutron counts due to high LLD settings, the neutron count rates are normalized to rate at 62 keV LLD setting based on the measured neutron response spectra. As shown in Fig. 5, the neutron count rate drops with an increased gamma-ray flux. At a gamma-ray flux of 1.37×10^6 photons/cm²s, a normalized neutron count rate of 0.95 with an LLD of 95 keV was obtained, a 5% reduction in neutron count rate. As the gamma-ray flux increases to 1.37×10^9 photons/cm²s, the normalized neutron count rate drops to 0.85 with an LLD of 182 keV, which corresponds to a 15% reduction in neutron count rate and a $\varepsilon_{int n}$ of 35% is maintained.

Fig. 5 Neutron count rates normalized to the one with no gamma-ray source present (62 keV LLD), measured with a $2 \times 2 \text{ mm}^2$ 50- μm -tall pillar detector. The LLDs are set to meet the requirement of $0.9 \leq \text{GARRn} \leq 1.1$.

We have demonstrated that our pillar structured thermal neutron detector based on a Si-¹⁰B composite can simultaneously achieve a high thermal neutron detection efficiency of 39% as well as a high n/γ discrimination of 8.5×10^5 in a high gamma-ray field of 10^9 photons/cm²s because of the small overall Si volume in the active region. While tuning the LLD to meet the gamma-ray insensitivity requirement of $0.9 \leq \text{GARRn} \leq 1.1$, a thermal neutron detection efficiency of 35% is maintained.

The authors would like to acknowledge Tim Graff and Cathy Reinhardt for their excellent support in the clean room fabrication. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract No. DE-AC52-07NA27344, LLNL-JRNL-641356. This work has been supported by the US Department of Homeland Security, Domestic Nuclear Detection Office, under competitively awarded IAA HSHQDC-08-X-00874. This support does not constitute an express or implied endorsement on the part of the government.

- 195 [1] R. T. Kouzes, "The ^3He supply problem," Pacific Northwest National Laboratory Report
196 PNNL-18388, 2009.
- 197 [2] W. J. McNeil, S. L. Bellinger, T. C. Unruh, E. L. Patterson, J. K. Shultis, and D. S.
198 McGregor, "Perforated diode fabrication for neutron detection," *IEEE Nucl. Sci. Symp.*
199 *Conf. Rec.* **6**, 3732-3735 (2006).
- 200 [3] D. S. McGregor, W. J. McNeil, S. L. Bellinger, T. C. Unruh, and J. K. Shultis,
201 "Microstructured semiconductor neutron detectors," *Nucl. Instr. and Meth. A* **608**, 125-131
202 (2009).
- 203 [4] A. M. Conway, T. F. Wang, N. Deo, C. L. Cheung, and R. J. Nikolic, "Numerical
204 simulations of pillar structured solid state thermal neutron detector: efficiency and gamma-
205 ray discrimination," *IEEE Trans. Nucl. Sci.* **56**, 2802-2807 (2009).
- 206 [5] R. J. Nikolic, A. M. Conway, C. E. Reinhardt, R. T. Graff, T. F. Wang, N. Deo, and C. L.
207 Cheung, "Pillar structured thermal neutron detector with 6:1 aspect ratio," *Appl. Phys. Lett.*
208 **93**, 133502 (2008).
- 209 [6] Q. Shao, R. P. Radev, A. M. Conway, L. F. Voss, T. F. Wang, R. J. Nikolic, N. Deo, and C.
210 L. Cheung, "Gamma-ray discrimination in pillar structured thermal neutron detectors," *Proc.*
211 *SPIE* **8358**, 83581N (2012).
- 212 [7] Q. Shao, L. F. Voss, A. M. Conway, R. J. Nikolic, M. A. Dar, and C. L. Cheung, "High
213 aspect ratio composite structures with 48.5% thermal neutron detection efficiency," *Appl.*
214 *Phys. Lett.* **102**, 063505 (2013).
- 215 [8] T. W. Crane and M. P. Baker, Chapter 13 "Neutron Detectors," *Passive Nondestructive*
216 *Assay of Nuclear Materials*, Doug Reilly, Norbert Ensslin, Hastings Smith Jr, and Sarah
217 Kreine, eds, (US Nuclear Regulatory Commission, 1991).
- 218 [9] K. -C. Huang, R. Dahal, J. J. -Q. Lu, A. Wertz, Y. Danon, and I. B. Bhat, "Scalable large-
219 area solid-state neutron detector with continuous p-n junction and extremely low leakage
220 current," *Nucl. Instr. and Meth. A* **763**, 260-265 (2014).
- 221 [10] K. -C. Huang, R. Dahal, J. J. -Q. Lu, Y. Danon, and I. B. Bhat, "High detection efficiency
222 micro-structured solid-state neutron detector with extremely low leakage current fabricated
223 with continuous p-n junction," *Appl. Phys. Lett.* **102**, 152107 (2013).

- [11] R. Dahal, K. C. Huang, J. Clinton, N. LiCausi, J. -Q. Lu, Y. Danon, and I. Bhat, "Self-powered micro-structured solid state neutron detector with very low leakage current and high efficiency," *Appl. Phys. Lett.* **100**, 243507 (2012).
- [12] S. L. Bellinger, R. G. Fronk, W. J. McNeil, T. J. Sobering, and D. S. McGregor, "Improved high efficiency stacked microstructured neutron detectors backfilled with nanoparticle ^6LiF ," *IEEE Trans. Nucl. Sci.* **59**, 167-173 (2012).
- [13] G. F. Knoll, *Radiation Detection and Measurement*, 3rd ed. (Wiley, New York, 2000).
- [14] J. W. Murphy, G. R. Kunnen, I. Mejia, M. A. Quevedo-Lopez, D. Allee, and B. Gnade, "Optimizing diode thickness for thin-film solid state thermal neutron detectors," *Appl. Phys. Lett.* **101**, 143506 (2012).
- [15] N. Deo, J. R. Brewer, C. E. Reinhardt, R. J. Nikolic, and C. L. Cheung, "Conformal filling of silicon micro-pillar platform with ^{10}B ," *J. Vac. Sci. Technol. B* **26**, 1309-1314 (2008).
- [16] ANSI/IEEE Std 301-1988, IEEE Standard Test Procedures for Amplifiers and Preamplifiers used with Detectors of Ionizing Radiation.
- [17] ANSI/IEEE Std 300-1988, IEEE Standard Test Procedures for Semiconductor Charged Particle Detectors.
- [18] ANSI 42.35-2006, American National Standard for Evaluation and Performance of Radiation Detection Portal Monitors for Use in Homeland Security. Technical Report. American Nuclear Standards Institute, Washington, D. C.
- [19] R. T. Kouzes, J. R. Ely, A. T. Lintereur, and D. L. Stephens, "Neutron detector gamma-ray insensitivity criteria," PNNL-18903 (2009).