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Contents lists available at ScienceDirect

Nuclear Instruments and Methods in
Physics Research Ajournal homepage: www.elsevier.com/locate/nimaBoron-coated straws as a replacement for ^3He -based neutron detectorsJeffrey L. Lacy*, Athanasios Athanasiades, Liang Sun, Christopher S. Martin,
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ARTICLE INFO

Available online 21 September 2010

Keywords:

Radiation portal monitoring
Long-range detection
Active interrogation

ABSTRACT

US and international government efforts to equip major seaports with large area neutron detectors, aimed to intercept the smuggling of nuclear materials, have precipitated a critical shortage of ^3He gas. It is estimated that the annual demand of ^3He for US security applications alone is more than the worldwide supply. This is strongly limiting the prospects of neutron science, safeguards, and other applications that rely heavily on ^3He -based detectors. Clearly, alternate neutron detection technologies that can support large sensitive areas, and have low gamma sensitivity and low cost must be developed.

We propose a low-cost technology based on long copper tubes (straws), coated on the inside with a thin layer of ^{10}B -enriched boron carbide ($^{10}\text{B}_4\text{C}$). In addition to the high abundance of boron on Earth and low cost of ^{10}B enrichment, the boron-coated straw (BCS) detector offers distinct advantages over conventional ^3He -based detectors, and alternate technologies such as $^{10}\text{BF}_3$ tubes and ^{10}B -coated rigid tubes. These include better distribution inside moderator assemblies, many-times faster electronic signals, no pressurization, improved gamma-ray rejection, no toxic or flammable gases, and ease of serviceability.

We present the performance of BCS detectors dispersed in a solid plastic moderator to address the need for portal monitoring. The design adopts the outer dimensions of currently deployed ^3He -based monitors, but takes advantage of the small BCS diameter to achieve a more uniform distribution of neutron converter throughout the moderating material. We show that approximately 63 BCS detectors, each 205 cm long, distributed inside the moderator, can match or exceed the detection efficiency of typical monitors fitted with a 5 cm diameter ^3He tube, 187 cm long, pressurized to 3 atm.

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1. Introduction

The diminishing inventory and minute natural abundance of ^3He gas on Earth necessitate the adoption of new technologies for the detection of neutrons, especially in homeland security applications, where large volume deployments requiring many liters of ^3He are required.

The only practical source of ^3He on Earth is achieved through production of tritium gas in specialized high flux reactors. Tritium decays to ^3He at a rate of 5.5% per year. Tritium was produced over the time frame from 1955 to 1988 for use as a critical ingredient of nuclear weapons. Production ceased in the United States in 1988 and is unlikely to resume, as there is currently an adequate supply to sustain the diminishing nuclear weapons inventory. The current US production rate of ^3He derived from the remaining tritium stock is estimated at 8000 l per year [1]. The US demand in recent years has been close to 70,000 l annually [1].

The US Department of Homeland Security (DHS) and Customs and Border Protection (CBP) plan to equip major US ports of entry with large area neutron detectors, in an effort to intercept the

smuggling of nuclear materials, potentially used in terrorist attacks. It is estimated that the annual demand of ^3He for US security applications alone is 22,000 l [2]. Clearly, alternate neutron detection technologies must be adopted, however, no attractive solutions have been successfully identified for large detectors that must have low gamma-ray sensitivity and low cost.

We propose a neutron detection technology based on long copper tubes ("straws"), 4 mm in diameter, coated on the inside with a thin layer of ^{10}B -enriched boron carbide ($^{10}\text{B}_4\text{C}$). Thermal neutrons captured in ^{10}B are converted into secondary particles, through the $^{10}\text{B}(n,\alpha)$ reaction. The reaction products, namely an alpha particle (α) and a lithium nucleus (^7Li) are emitted isotropically from the point of neutron capture in exactly opposite directions and, in the case of the dominant excited state, with kinetic energies of 1.47 and 0.84 MeV, respectively. Straw detectors can be easily and economically manufactured in large quantities, in a manner that guarantees a $^{10}\text{B}_4\text{C}$ lining that is pure, uniform, and optimally thin. By making the boron carbide layer thin, typically 1 μm , one or the other of the two charged particles (whichever is directed inward) has a high probability to escape the wall and ionize the gas contained within the straw. The detection efficiency achieved is several times higher than that of standard boron-coated counters.

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We have previously published on the BCS detection capabilities, fabrication, and development of prototypes for various applications [3–7]. Here we investigate the use of the BCS detectors in radiation portal monitors.

2. Methods

³He tubes enclosed in moderator boxes are a standard solution in radiation portal monitor (RPM) applications, for the screening of cargo at ports of entry, and other industrial applications. In a standard design, either one or two ³He tubes, 187 cm long, pressurized to 3 atm, are enclosed in a high-density polyethylene (HDPE) box, as shown in Fig. 1. The box has outer dimensions of 30.5 × 12.7 × 215 cm³ (W × D × H), and is assembled from separate pieces, with an internal opening that accommodates the tubes. The box height is 215 cm. This assembly is specified [8] to have a sensitivity of at least 20 cps for a ²⁵²Cf source emitting 20,000 neutrons/s, placed at 2 m from the tube center. For a two-tube assembly, the count rate is specified at 32 cps.

RPM installations consist at a minimum of 2 such panels, and typical installations employ four panels, two on either side of a drive-through lane. It is estimated that 1100 RPM installations have been deployed [2], for a total ³He volume of at least 44,000 l.

The BCS detector can be manufactured in large volumes to replace ³He-based RPM designs, and support widespread deployment. An RPM design based on BCS detectors is presented in Fig. 2, maintaining the outer dimensions of the HDPE box of Fig. 1 for a direct performance comparison. Due to its small diameter and high segmentation, the BCS can be uniformly distributed inside the moderating material in a manner that maximizes sensitivity.

2.1. BCS-based RPM design simulation

The response of the BCS-based RPM has been determined in Monte Carlo simulations, implemented in MCNP5. The BCS detectors were 4 mm in diameter, 205 cm long, and were lined with 1 μm thick ¹⁰B₄C. The source was placed 2 m from the moderator front surface, and emitted 20,000 neutrons/s isotropically and with energies as specified by the built-in MCNP5 fission spectrum for ²⁵²Cf. The source was surrounded with 0.5 cm thick lead, and 2.5 cm thick HDPE. Neutrons were allowed to thermalize and be reflected back out from the concrete ground, and from a steel box surrounding the detector, as illustrated in Fig. 3.

Computed count rates are plotted in Fig. 4 as a function of the number of straw detectors. The 63-straw design performance is 20 cps, matching that of the one-³He-tube RPM of Fig. 1, and the

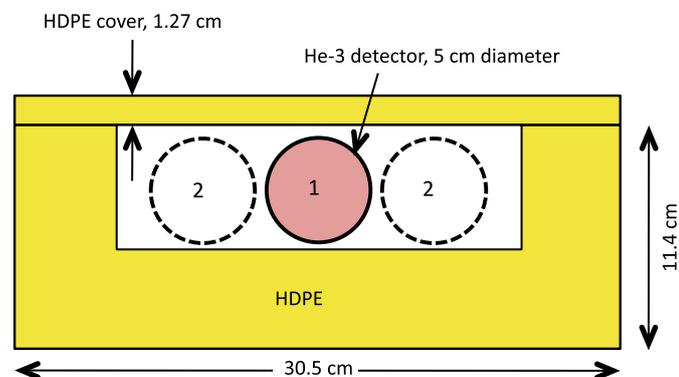


Fig. 1. Typical design and dimensions of a radiation portal monitor (RPM), with a single ³He tube (dashed circles show possible tube locations in the two-tube design). The high-density polyethylene (HDPE) box has a height of 215 cm. The tube is pressurized to 3 atm.

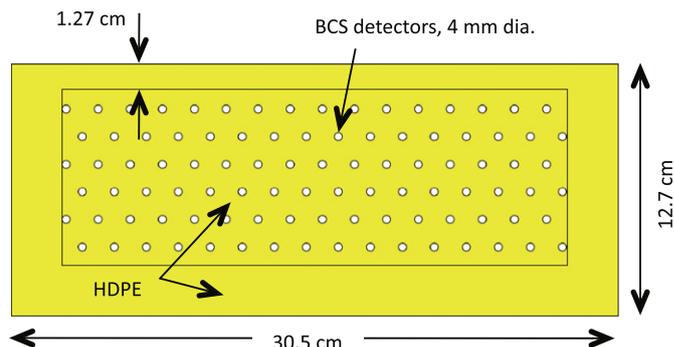


Fig. 2. Radiation portal monitor (RPM) design based on boron-coated straw (BCS) detectors. The BCS detectors (96 shown here) are uniformly distributed within the central 25.4 × 8.89 cm² area of high-density polyethylene (HDPE). The outer dimensions are those typically encountered in ³He-based designs (see Fig. 1). Each straw detector is 205 cm long.

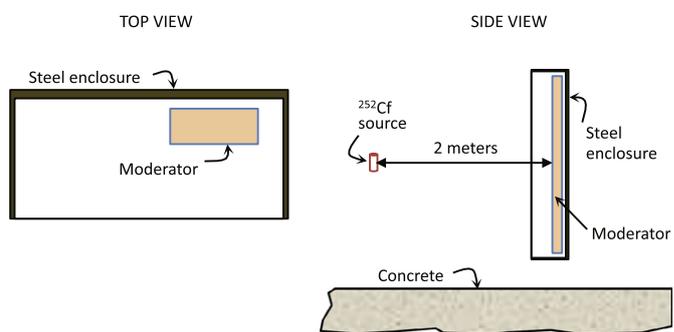


Fig. 3. Top-view and side-view illustrations of geometry setup used in Monte Carlo simulations of the proposed BCS-based detector for portal monitors.

111-straw design matches the performance of the 2-tube RPM. A significant improvement in sensitivity can be obtained with higher straw densities. At densities higher than 200 straws, the performance benefit is small, indicating that the moderation of high-energy neutrons is not adequate. In the range beyond 800 straws, an increase in the number of straws reduces the detection efficiency.

2.2. Prototype testing

A prototype portal monitor was built, based on a design that distributes BCS detectors inside a solid block of high-density polyethylene (HDPE), as described earlier. The outer dimensions of the HDPE block were 30.5 × 12.7 × 215 cm³ (W × D × H). The HDPE block, shown in Fig. 5, has a total of 171 holes, with a pitch of about 10 mm, and can accommodate an equal number of BCS detectors. The BCS detectors were 4 mm in diameter, 200 cm long, and were lined with ¹⁰B₄C.

A total of either 63 or 85 BCS detectors were used for the measurements presented here. The detectors occupied every other hole in the HDPE block, as shown in Fig. 5. The straw walls (cathodes) of all detectors were connected together and grounded, using an aluminum plate, shown in Fig. 5. The signals were read out by connecting all anodes to a single charge sensitive preamplifier (Canberra, model 2006). An external shaper (Canberra model 2022, 1 μs shaping time) and multichannel analyzer (Amptek MCA-8000) were used. The detectors were biased to 700 V through a 34 nF coupling capacitor.

Measurements were conducted with two different ²⁵²Cf sources, both purchased from Frontier Technology (FT). The sources were measured by FT, and found to be 6.03 ± 0.18 μg on 11/17/2008 for the larger source, and 1.40 ± 0.042 μg on 2/21/2002 for the smaller

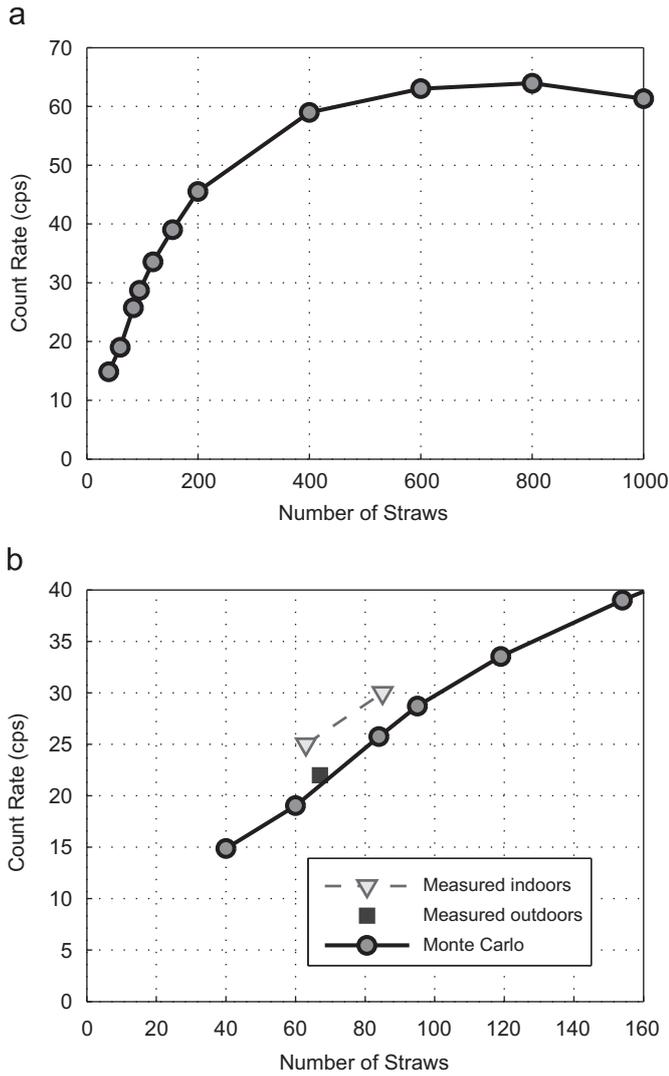


Fig. 4. (a) Predicted count rate in BCS-based detector design of Fig. 2, for a ^{252}Cf source, emitting 20,000 neutrons/s at 2 m, as a function of the number of straws. (b) Same as (a), including measurements collected with the prototype monitor of Fig. 5. The detector was oriented horizontally for the indoors measurements, and vertically, in front of a steel plate, for the outdoors measurements.

source. The error in the FT measurements was $\pm 3\%$. On the day of the measurements presented here, the source sizes were 4.11 ± 0.12 and 0.164 ± 0.0049 μg , respectively. The corresponding neutron emission rates are 9.45×10^6 and 0.377×10^6 neutrons/s, respectively. In all measurements, the sources were placed inside a pig that surrounded the source with 0.5 cm thick lead, and 2.5 cm thick polyethylene. The portal monitor was lying on a laboratory bench, oriented with its long axis parallel to the ground, as shown in Fig. 5, and with its 30 cm long side facing the source, which was placed 2 m away from the monitor side facing it. Both the source and the monitor center were 110 cm above the concrete floor. The source was supported on a tripod.

A 10 mCi ^{137}Cs gamma source was used to test the gamma rejection capability of the prototype portal monitor. The portal monitor was positioned in the same manner as described above, for the neutron measurements. The gamma source was supported on a tripod and placed 61 cm from the front side of the monitor, and 1 m above the concrete floor. The exposure rate measured at the monitor was 10 mR/h.

Additional measurements were conducted outdoors, with the detector populated with 67 straws, and its long axis oriented vertically. The detector center was located 1.5 m above the ground. A 25.4 mm thick steel plate was positioned directly behind the moderator block. The ^{252}Cf source was also placed 1.5 m above the ground. Other parameters were set as described above for the indoor measurement.

3. Results

The neutron count rate was initially recorded in the absence of sources. Over a period of 4278 s, a total of 7683 counts were recorded, giving a background rate of 1.80 cps. The energy spectrum of background counts is shown in Fig. 6.

The net count rate recorded with the large ^{252}Cf source and 85 straws was 13,714 cps. The net rate recorded with the small source was 566 cps. The corresponding sensitivities, obtained by dividing the count rate by the source amount, were 3.34 ± 0.10 and 3.45 ± 0.10 cps/ng, respectively. The error in these measurements is due to the uncertainty in the neutron source size, discussed earlier ($\pm 3\%$). The net rate recorded with 63 straws was 2.86 cps/ng. The US government requirement for portal monitors is 2.5 cps/ng [9].

The above rates can be scaled down to the size of a standard ^{252}Cf source that emits 20,000 neutrons/s. The results are 29.0 ± 0.87 cps (large source) and 30.0 ± 0.90 cps (small source)

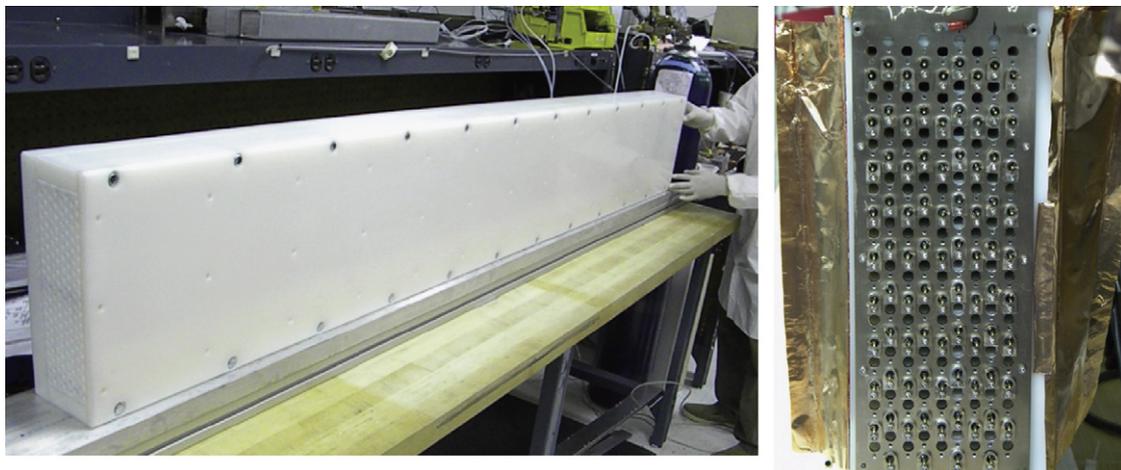


Fig. 5. Prototype BCS-based portal detector.

Table 1
Count rates measured with ^{252}Cf sources at 2 m.

Source size	Background rate (cps)	Net counts	Collection time (s)	Net count rate (cps)	Sensitivity (cps/ng)	Net count rate for standard source ^a (cps)
85 STRAWS						
Large ^{252}Cf source ^b	1.80	1,398,792	102	13,714	3.34 ± 0.10	29.0 ± 0.87
Small ^{252}Cf source ^c	1.80	283,470	501	566	3.45 ± 0.10	30.0 ± 0.90
63 STRAWS						
Small ^{252}Cf source ^c	1.40	40,755	86.9	469	2.86 ± 0.09	24.9 ± 0.75

^a Standard source emits 20,000 neutrons/s.

^b Large source is $4.11 \pm 0.12 \mu\text{g}$.

^c Small source is $0.164 \pm 0.0049 \mu\text{g}$.

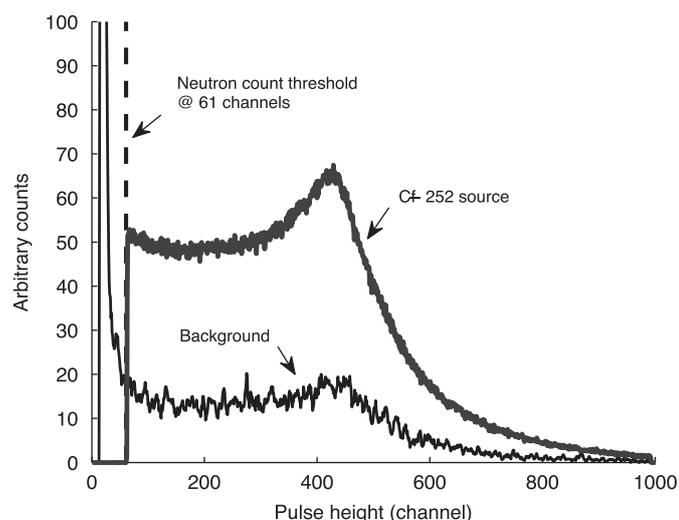


Fig. 6. Pulse height spectra of background counts collected in the prototype detector, and spectra collected with the ^{252}Cf source at 2 m.

for the 85-straw configuration, and 24.9 ± 0.75 cps for the 63-straw configuration. The results are summarized in Table 1 and plotted in Fig. 4(b). The pulse height spectra corresponding to this measurement (85 straws and large source) are shown in Fig. 6.

By comparison, the corresponding performance of ^3He -based RPM's deployed by the US government is at least 20 cps in designs that employ a single ^3He tube, and at least 32 cps in designs that employ 2 tubes [8].

The rate measured with the 67-straw detector outdoors, and in front of a steel plate, was 2.53 ± 0.08 cps/ng, corresponding to 22.0 ± 0.66 cps for the standard source.

Neutron counts collected with a 10 mR/h gamma exposure, over a 3708 s time interval, totaled 6439. This corresponds to a neutron background rate of 1.74 cps. Another collection was done with a 50 mR/h exposure, resulting in a rate of 1.80 cps. The pulse height spectra are shown in Fig. 7.

4. Discussion

The rates predicted in the simulation of Fig. 4 were 26 cps, for the 85-straw design, and 20 cps for the 63-straw design, about 13% and 20% lower than the measured rates, respectively. These rates are indicated in Fig. 4 for a direct comparison with the predicted rates. We believe the difference is due to reflections from various laboratory structures and furniture surrounding the detector, and due to its horizontal orientation. The outdoors measurement of 22 cps, also indicated in the figure, shows better agreement with the simulation.

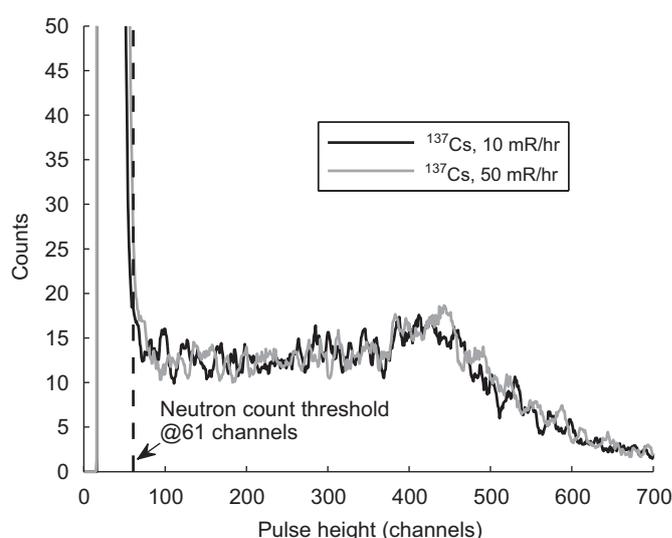


Fig. 7. Pulse height spectra collected in the prototype detector under different gamma exposure rates.

Future measurements will be designed to more accurately represent the response of the detector in actual outdoors RPM settings, where the detector/moderator assembly is installed inside a steel enclosure (as presented in the Monte Carlo simulation), alongside other detectors. Nevertheless, we showed that the detector design of Fig. 2, populated with 63 straw detectors, could match or exceed the performance of the ^3He -based design. The amount of ^{10}B neutron converter present in the 63-straw assembly is 2.97 g, or 0.297 moles of ^{10}B . The amount of ^3He present in the one-tube RPM design discussed previously is 1.38 g, or 0.458 moles of ^3He . Accounting for the neutron absorption cross-sections of the two isotopes (3835 barn for ^{10}B and 5333 barn for ^3He), we conclude that with ^{10}B we use 2.1 times less neutron converter than ^3He , in this application.

This benefit is due to the optimization of moderator material surrounding the detectors. A large number of detectors with moderator in-between allows more neutrons to be captured following thermalization, since fewer neutrons are absorbed in the plastic medium between the detectors. With the bulky ^3He design, neutrons must travel long distances in plastic after reaching thermal energies prior to encountering the detection medium.

4.1. Other applications

Neutron detectors based on pressurized ^3He gas are commonly employed in the detection of fission neutrons in active interroga-

tion setups. However, the recovery time of ^3He tubes is too long, typically ~ 2 ms, and prevents efficient counting of fission neutrons generated with fast linac pulses. The ionization produced in the detector during the brief linac pulse must be cleared (i.e., positive ions must be collected at the cathode) before the detector is able to count neutrons.

The small size, low pressure, and free choice of counting gas in BCS detectors translate to fast recovery times that can be as low as $20\ \mu\text{s}$. A faster detector recovery can carry profound benefits for this application. Prompt neutrons can thermalize and be detected as early as $30\ \mu\text{s}$ [10], following the linac pulse, with a yield per fission about 150 times higher than that of delayed neutrons. Most prompt neutrons will die away within the first millisecond [10], thus, a detector that requires a longer recovery time will not count them.

The BCS detector has been additionally proposed for long-range detection and localization of hidden neutron and gamma sources, in homeland security applications [7]. Straw detectors can be stacked to form a large area ($1\ \text{m}^2$ or higher) panel that is lightweight and safe to transport. A limited-scale ($0.5\ \text{m}^2$) prototype incorporating eleven detector modules has been constructed. Each module consisted of a 5×10 close-packed array of copper straws, each 4 mm in diameter, 100 cm long, and lined with a nominal $1\ \mu\text{m}$ thick coating of enriched $^{10}\text{B}_4\text{C}$. A number of tests were performed successfully, including temperature cycling and mechanical vibration.

5. Conclusion

It is imperative that ^3He -free neutron detection technologies be adopted, to overcome limitations imposed by a depleting ^3He supply and sharply rising neutron detection demand. The ^{10}B medium is abundant and easy to produce, making possible the extensive deployment of neutron detectors worldwide for the detection of nuclear threats.

The boron-coated straw (BCS) offers distinct advantages over conventional ^3He -based neutron detectors, including free choice of counting gas, that can be replaced as needed, no pressurization, low weight, faster signals for better immunity to low-frequency

microphonic noise, faster recovery time in gamma activation applications and good gamma discrimination with simple pulse height discrimination.

The proposed technology can have a significant impact in security applications, where large sensitive volumes are required. The BCS can be readily applied to portal monitoring, freeing up large amounts of ^3He gas that can be redistributed to other applications, such as neutron science and nuclear safeguards.

Funding

This work was funded by the Domestic Nuclear Detection Office (DNDO).

References

- [1] W. Brinkman, Testimony before US House Subcommittee on Investigations and Oversight, Committee on Science and Technology, April 22, 2010, available online at: http://democrats.science.house.gov/Media/file/Commdocs/hearings/2010/Oversight/22apr/Brinkman_Testimony.pdf.
- [2] R.T. Kouzes, in: "The ^3He Supply Problem", PNNL-18388, Pacific Northwest National Laboratory, Richland, Washington, 2009.
- [3] A. Athanasiades, N.N. Shehad, C.S. Martin, L. Sun, J.L. Lacy, Straw detector for high rate, high resolution neutron imaging, in: IEEE Nuclear Science Symposium Conference Record, vol. 2, 2005, pp. 623–627.
- [4] J.L. Lacy, A. Athanasiades, N.N. Shehad, C.S. Martin, L. Sun, Performance of 1 meter straw detector for high rate neutron imaging, in: IEEE Nuclear Science Symposium Conference Record, vol. 1, 2006, pp. 20–26.
- [5] A. Athanasiades, N.N. Shehad, L. Sun, T.D. Lyons, C.S. Martin, L. Bu, J.L. Lacy High sensitivity portable neutron detector for fissile materials detection, in: IEEE Nuclear Science Symposium Conference Record, vol. 2, 2005, pp. 1009–1013.
- [6] J.L. Lacy, A. Athanasiades, C.S. Martin, L. Sun, J.W. Anderson, T.D. Lyons, Long range neutron–gamma point source detection and imaging using unique rotating detector, in: IEEE Nuclear Science Symposium Conference Record, vol. 1, 2007, pp. 185–191.
- [7] J.L. Lacy, A. Athanasiades, L. Sun, C.S. Martin, G.J. Vazquez-Flores, Boron coated straw detectors as a replacement for ^3He , in: IEEE Nuclear Science Symposium Conference Record, 2009, pp. 119–125.
- [8] SAIC specification sheet, Exploranium™ AT-980 Radiation Portal Monitor.
- [9] D. Stromswold, J. Ely, R. Kouzes, J. Schweppe, in: "Specifications for Radiation Portal Monitor Systems Revision 6.7", PIET-43741-TM-017, Pacific Northwest National Laboratory, Richland, Washington, 2003.
- [10] T. Gozani, Active nondestructive assay of nuclear materials: principles and applications, Technical report, NUREG/CR-0602; SAI-MLM-2585, 1981.