

Recent progress in the commercialization of the Li Foil multi-wire proportional counter neutron detectors

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ABSTRACT

The scarcity and rising cost of ³He gas has introduced a push in alternative neutron detector technologies. The Li foil multi-wire proportional counter (Li Foil MWPC) neutron detectors have shown promise as a ³He replacement technology. Large, gas-filled proportional counters with five layers of 75 μm thick ⁶Li foils have been built and characterized, yielding over 55% thermal neutron detection efficiency, with the possibility of increasing the efficiency above 70% with ten 96% enriched ⁶Li foil layers. Most recently, a backpack radiation detector (BRD) was fabricated, equipped with four Li Foil MWPC devices. The neutron sensitivity-to-weight ratio of the Li Foil MWPC devices was first optimized using MCNP6 by simulating the angular response to an ANSI-moderated-²⁵²Cf source positioned 1.5 m away from the devices. The simulated neutron sensitivity results were compared and benchmarked to that of the commercially-available Thermo-Fisher PackEye v. 1.0 that contains two 14 in. long, 2 in. diameter, 2.5 atm ³He tubes. The experimental efforts studied the performance of the Li Foil MWPCs in the backpack instrument configuration. Fabrication advancements were implemented to the Li Foil MWPC devices to increase the device manufacturability and improve the commercialization capabilities. These advancements included weight reduction, new hermetic sealing techniques, optimizing active area, and anode wire cost reduction. The Li Foil MWPC BRD was evaluated as a part of a PNNL test campaign for neutron sensitivity where 0.36 cps/ng of ²⁵²Cf at 1.5 m was measured. The gamma-ray rejection ratio (GRR) of the BRD was also evaluated, yielding a measured value of 3.1×10^{-8} for a ¹³⁷Cs exposure rate of 50 mR/Hr. Further evaluations were performed under the PNNL test campaign including detector response to neutron sources of ²⁵²Cf, AmBe, AmLi and WGPu, detector angular response, and the gamma absolute rejection ratio in the presence of neutrons (GARRn).

1. Introduction

Li foil multi-wire proportional counter (Li Foil MWPC) neutron detectors utilizing the suspension of ⁶Li-foil sheets between anode wire banks have been investigated at Kansas State University (KSU) since 2009 (McGregor et al., 2012, 2013). The Li Foil MWPC neutron detectors are able to achieve high neutron detection efficiencies because the Li foil sheets are suspended and stacked one behind another, sandwiched between anode wire banks (Nelson et al., 2012a). The ranges of the triton and alpha particle reaction products from the ⁶Li(n, α)³H reaction are 133 μm and 23 μm, respectively, thus, the summed range of the reaction products is 156 μm (Nelson et al., 2012a). Due to the advancements in the lithium-ion battery industry, ⁶Li foils can be fabricated as thin as 55 μm. For sufficiently thin ⁶Li foils, the summed

range of the reaction products is greater than the thickness of the Li foil sheet, and, therefore, one or both reaction products can escape the Li foil sheet simultaneously. Consequently, both reaction products are concurrently capable of entering into the ionizing gas volume causing relatively large-amplitude pulses compared to simple Li-coated devices. This attribute results in a pulse-height spectrum where a valley appears between the electronic noise and the main neutron-induced features in the spectrum. The thickness and quantity of Li foils directly affects the neutron detection efficiency. Thus, the maximum neutron detection efficiency occurs at an optimal foil thickness depending on the number of foils. Specifically, as mentioned above, the detectors constructed to date contain five layers of 75-μm thick foil, the optimized thickness for maximum neutron detection efficiency for a five foil system. Further, the gamma-ray rejection ratio (GRR) of these detectors is typically

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1.0×10^{-8} or better for ^{137}Cs exposure rates of 20–50 mR/h, and is achieved through pulse-height discrimination methods (setting the threshold, or lower level discriminator (LLD), in the spectral valley) (Nelson et al., 2015, 2014). The LLD is set towards the higher-energy range of the spectral valley in the pulse-height spectrum, which eliminates essentially all pulses from gamma-ray interactions. This valley does not appear in pulse-height spectra obtained with coated Li foil detectors because only one of the reaction products is able to enter the ionizing gas volume (McGregor et al., 2003). The other reaction product, emitted in the opposite direction, typically deposits its energy within the Li coating or the chamber wall. Consequently, this significantly reduces the neutron detection efficiency and the ability to easily discriminate gamma-ray radiation without sacrificing significant neutron-induced counts.

Portable (i.e. backpack) detection systems containing Li Foil MWPCs have been fabricated in the past. These backpack radiation detectors (BRDs) contained Li Foil MWPCs with five layers of 75- μm thick ^6Li foil. The simulated intrinsic thermal neutron detection efficiency of these devices was 55% (Nelson et al., 2012a, 2014, 2012b). The first portable unit weighed 24 bs (10.9 kg) and was 4.75 in. (12 cm) thick. The second portable unit contained three Li Foil MWPCs, each with outer dimensions of 2 in. x 5 in. x 15 in. These were some of the first iterations of a backpack style Li Foil MWPC. The following studies focus on adding new capabilities to-, expanding existing capabilities on-, and increasing the technology readiness of- the prototype Li Foil MWPC. In doing so, a BRD was fabricated and underwent a series of evaluations at Pacific Northwest National Laboratory (PNNL). The BRD design followed the specifications outlined in the ANSI N 42.53 standards. Some of the most relevant advancements to the Li Foil MWPC include improvements to the design of the detector enclosure and anode wires, refined fabrication methods, and the implementation of hermetic sealing procedures.

2. Modeling and initial design

Simulations were performed in order to evaluate and compare the neutron-detection capabilities of the Li Foil MWPCs. The following modeling results provided insight regarding the functionality of the Li Foil MWPC design. This allowed for optimization of the design to adhere to the desired design criteria pertaining particularly to neutron sensitivity and BRD weight.

Three generations of ^6Li foil BRD device designs were simulated, which led to the final design used in the BRD build. This final design was compared to the Thermo Fisher PackEye v. 1.0, which includes two 14 in. long, 2 in. diameter, 2.5 atm ^3He tubes. The generation-1 design did not meet the weight specification for BRDs, as specified by ANSI N 42.53. The design of generations 2 and 3 were modified primarily to reduce weight while maintaining/improving the neutron sensitivity of the BRD. Generation-2 was modified by decreasing the total number of devices from four to two, decreasing the overall length of each device from 40 cm to 37 cm, and incorporating an additional suspended 75 μm thick ^6Li foil into each device. The decrease in the number of devices and device length resulted in less material being used for each device enclosure, effectively reducing the weight of each device. Thus, the primary modification to the device design, as shown in the generation-3 (GEN 3) ^6Li foil BRD device design (Fig. 3), was that the wall thickness of the device enclosure was reduced from 1/8 in. to 1/16 in. aluminum to further reduce the weight of each device. Furthermore, an additional suspended 75 μm thick ^6Li foil was also incorporated into each device of the GEN 3 ^6Li foil BRD device design. The simulated neutron-sensitivity responses to ^{252}Cf in the horizontal source plane for the GEN 3 Li Foil MWPC with 1/4 inch-thick high density polyethylene (HDPE) moderator, 1/8 in.-thick HDPE moderator, and no HDPE moderator surrounding the BRD are shown in Fig. 1. The neutron sensitivity of the Thermo Fisher PackEye v. 1.0 was also simulated for comparison purposes. The configuration of the detectors, relative to the HDPE moderator arranged in the BRD, is shown in Fig. 2. An illustration of the GEN 3 detector final

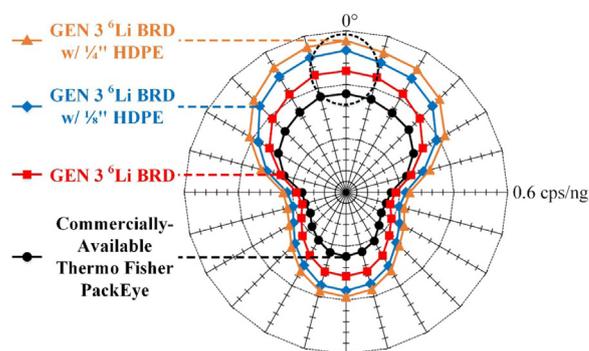


Fig. 1. Angular neutron-sensitivity response of the GEN 3 Li Foil MWPC design iterations compared to the Thermo Fisher PackEye v. 1.0. The neutron-sensitivity responses were simulated for a bare ^{252}Cf source positioned within the horizontal source plane in increments of 15° .

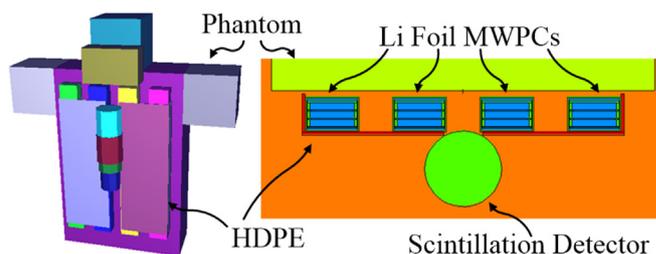


Fig. 2. The BRD arranged against a phantom (left), and the detector/moderator configuration with 1/8 in. thick high-density polyethylene (HDPE) (right). A scintillation detector was also modeled with the BRD because it will be included in future BRD builds.

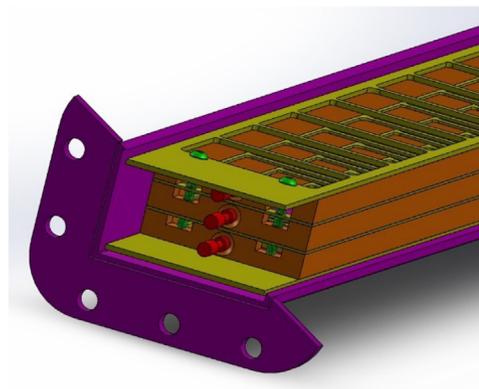


Fig. 3. The GEN 3 design of the Li Foil BRD.

iteration, equipped with 2 suspended foils and 2 wall foils, is shown in Fig. 3.

3. Experimental procedures

A number of Li Foil MWPC advancements were studied and implemented in the latest Li Foil MWPC design, including anode wire manufacturing procedures, detector containment material selection, and manufacturing methods. The following sections will elaborate on these advancements, and discuss the BRD that was fabricated and sent to PNNL for further evaluations.

3.1. Detector advancements

Commercialization of the Li Foil MWPC is the primary focus of this study, and therefore, the manufacturing methods to fabricate these



Fig. 4. An example of lithium laminated onto nickel-plated aluminum using an Apache laminator.

devices were explored to produce detectors relatively rapidly. One of these efforts was determining a fast and rugged method for suspending the Li foil. Lithium is very reactive, and, therefore, selecting materials that can come into contact with lithium without the lithium reacting can be difficult. Aluminum is a top choice for fabricating these devices given its relatively low density. However, when lithium comes into contact with bare aluminum (native oxide), lithium will react with the native oxide over time. One method for preventing the slow reaction between lithium and the native aluminum oxide is to apply a nickel plating to the surfaces of the aluminum component. Lithium does not react with the nickel native oxide. A 500 micro-inch layer of nickel was coated on all frames used in fabricating Li Foil MWPCs. Lithium was directly laminated to nickel-plated aluminum frames using a commercially-available APACHE laminator. The strip of lithium foil was placed over the top of the nickel-plated aluminum frame, then sandwiched between Teflon™ sheets, and the entire assembly was fed through the laminator. The resulting laminated Li foil layer on the nickel-plated aluminum frame is shown in Fig. 4. The adhesion between the Li foil and the nickel plated frame is such that the lithium rips when attempting to remove the foil from the frame. To remove any residual lithium from the frames, in the case of rebuilding a detector, the frames are placed in deionized water to convert the lithium to lithium hydroxide, and, thereby cleaning the frame of all lithium. The lithium hydroxide can be used to make other lithium compounds, such as lithium fluoride, for other lithium-based neutron detector technologies (Bellinger et al., 2012).

Anode wire manufacturing was also explored for mass-production and commercialization capabilities. The goal was to fabricate a rugged and robust wire that did not suffer from microphonic noise problems, and could be fabricated with off-the-shelf materials. Additionally, all materials chosen were low-outgassing and compatible with lithium to prevent contamination issues. The Li Foil MWPC anode was fabricated using 200 μm diameter stainless steel wire. The wire was fixed into place at one end of the anode frame by soldering the wire to a zirconia ferrule with an alumina washer, an item used in the fiber optics industry. The other end of the wire was strung through another zirconia ferrule at the other end of the frame with an alumina washer, vibration damping spring, and alumina insulation washer. The anode wire was attached to a weight that pulled tension on the wire, and the wire was

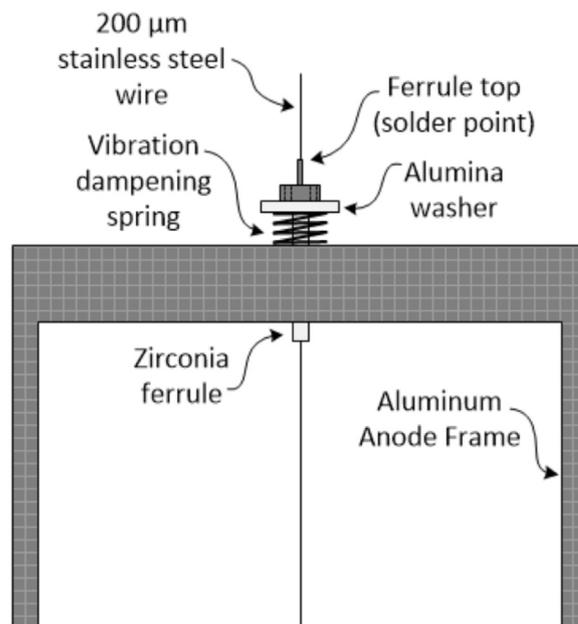


Fig. 5. A sketch of the top portion of the anode wire configuration used in the Li Foil MWPC. The use of a zirconia ferrule is the key component that combined an insulated region around anode wire and having a direct solder point at the top of the ferrule where the wire could be soldered into place while under tension.

soldered into the ferrule connection to complete the anode wire fabrication. The anode wire, positioned within an anode frame, is shown in Fig. 5.

Additional advancements to the Li Foil MWPC included reducing the weight of the detector in multiple ways. First, the containment tanks were manufactured from custom extruded aluminum tubing with a 1/16 in. side wall. Li Foil MWPCs have been built from enclosures with wall thicknesses up to 1/4 in. The reduction of the wall thickness of the containment tank yielded a reduction in weight, therefore the GEN 3 design resulted in the overall weight of the BRD being below the weight requirement specified in the ANSI N 42.53 standard. The detector weight was also reduced by adjusting the design of the lid and gas-backfill seals. Previously Li Foil MWPC designs relied on a bolt-on flange and a quarter-turn valve, which are components that add unnecessary weight to the device design. As a result, laser welding was explored in an attempt to remove the bolt-on flange, which performed very well. However, unfortunately, laser welding is costly. Therefore, other methods for lid seals were evaluated, such as a press-fit lid with a low-outgassing epoxy sealant. Additionally, in an effort to remove the bulky and heavy quarter-turn valve from the device, pinch-off tube sealing methods were explored as well. A copper tube was flared at one end and attached to the tank lid using a low-outgassing sealant, such as Torr Seal®. The copper tube was then pinched off using a set of pinch-off pliers manufactured by CHA Industries, Inc. Leak checks and lifetime evaluations are currently underway and these weight-reduction methods are going to be further explored in the phase II efforts of the project.

3.2. Backpack radiation detector (BRD) build

Multiple GEN 3 Li Foil MWPC devices were fabricated for a BRD. All fabrication procedures and lithium laminations were conducted within a dry (< 1.0 ppm water), argon environment (< 1.0 ppm oxygen) contained in a glove box. Devices were sealed using a bolt-on flange with a Viton® gasket to make the final containment seal, and closed off at the gas feed-through using a quarter turn valve. The sealed detector was then removed from the glove box, and was connected to a vacuum

station. The detector was evacuated and a bake-out procedure was performed overnight with the detector maintained under rough vacuum. The following day, the detector was pressurized with ultra-high purity (UHP) argon to 10 psig. Other backfill gases, such as P-10 proportional gas (90% argon, 10% methane), are prone to reacting with the lithium foil, leading to warping of the foils. Eventually, the warped foils can collide with anode wires causing the device to malfunction. Hydrocarbons, such as methane that is a common quench gas in gas-filled proportional counter detectors, gradually dissociate over time when used as a proportional gas. The gradual dissociation of the hydrocarbons can lead to the formation of lithium carbides as the dissociated carbon molecules slowly react with the lithium. The Li Foil MWPC was operated at 600 V, a voltage where the gas multiplication factor is relatively low, thereby making it possible to use a pure noble gas, such as argon, without the addition of a quench gas (Sipilä, 1977). Other materials consisting of hydrocarbons, such as Viton® and other plastics, also must be avoided inside the detector containments because these hydrocarbon sources can also lead to lithium contamination problems.¹ Illustrated in Fig. 2 is the configuration of four Li Foil MWPC devices that were connected together in series. The Li Foil MWPCs were placed in a backpack with 1/8-in. thick of HDPE surrounding the detectors. The final BRD is shown in Fig. 6 mounted to a human phantom for preliminary radiation sensitivity testing at Kansas State University (KSU). The preliminary radiation sensitivity measurements were conducted prior to the PNNL test campaign.



Fig. 6. The final Li Foil BRD that was fabricated at KSU and mounted to a human phantom for preliminary radiation-sensitivity testing prior to the PNNL testing campaign.

4. Results and discussion

The neutron sensitivity of the Li Foil MWPC BRD was evaluated using a ²⁵²Cf positioned at a distance of 1.5 m. Gamma-ray sensitivity measurements were performed using a ¹³⁷Cs gamma-ray source with an exposure rate of 50 mR/Hr. The neutron and gamma-ray sensitivity measurements were conducted prior to the PNNL testing campaign. Radiation sensitivity measurements were performed at KSU in a 24 ft x 15.1 ft x 13.5 ft concrete room. The measured net count rate from the ²⁵²Cf source at 1.5 m was 1.03 cps/ng. The same ²⁵²Cf testing scenario was also modeled using MCNP6 that yielded a predicted count rate of 1.06 cps/ng. A GRR of 3.1×10^{-8} was measured for a ¹³⁷Cs exposure rate of 50 mR/Hr. The spectra from these measurements and the modeled test scenario are shown in Fig. 7. Following preliminary radiation sensitivity measurements, the BRD was sent to PNNL for a test campaign. Initial neutron sensitivity testing at PNNL, conducted using a ²⁵²Cf source, yielded a net count rate of 0.36 cps/ng, which agreed with an MCNP model that included approximate dimensions and materials in the PNNL test scenario. The high count rate measured at KSU was largely due to neutron-albedo contributions from the surrounding concrete walls.

The Li Foil MWPC BRD underwent a variety of radiation-sensitivity evaluations at PNNL. The first evaluations involved measuring the neutron sensitivity of the BRD to separate sources of ²⁵²Cf, AmBe, WGPu, and AmLi at varying neutron fluence. The intrinsic neutron detection efficiency of the Li Foil MWPC BRD for these sources and fluences is shown in Fig. 8 and tabulated in Table 1. The measured intrinsic neutron detection efficiency was approximately 4–8% at each neutron fluence. An increased sensitivity to AmLi was observed likely due to the softer spectrum emitted from AmLi compared to that of ²⁵²Cf, WGPu and AmBe (Kouzes et al., 2008). Next, the angular neutron-sensitivity response of the Li Foil MWPC BRD was measured relative to the front of the detector face (0° position) with a 50 μ Ci bare-²⁵²Cf

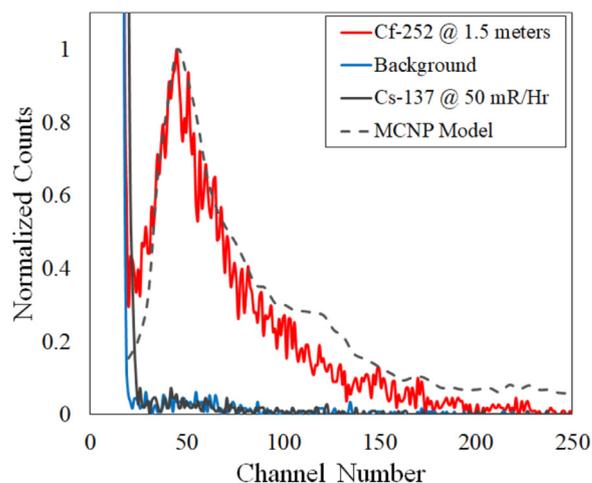


Fig. 7. The Li Foil BRD response to ²⁵²Cf at a distance of 1.5 m, relative to a background measurement, compared to the predicted response from MCNP. The response of the BRD to a ¹³⁷Cs exposure rate of 50 mR/Hr is also shown.

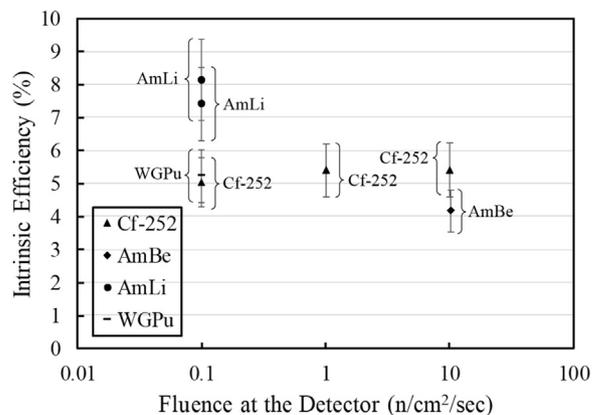


Fig. 8. The intrinsic neutron detection efficiency of the Li Foil BRD as a function of the neutron fluence from separate neutron sources of ²⁵²Cf, AmBe, WGPu, and AmLi sources.

¹ Although GEN 3 devices were produced in this study containing a Viton® gasket, efforts will be made to remove this high outgassing gasket in future builds. Not all devices malfunction as a result of hydrocarbon contamination, although the probability of device malfunction is higher when using hydrocarbon-based gases and detector components.

Table 1
Tabulated intrinsic neutron detection efficiency of the Li Foil BRD as a function of the neutron fluence from ²⁵²Cf, AmBe, WGPu, and AmLi sources.

Neutron source	Neutron fluence (n/cm ² /s)	Absolute efficiency (%)	Error (%)
²⁵² Cf	0.1	5.04	0.76
²⁵² Cf	1	5.4	0.81
²⁵² Cf	10	5.41	0.81
AmBe	10.2	4.17	0.63
AmLi	0.1	7.41	1.12
AmLi	0.1	8.14	1.23
WGPu	0.1	5.23	0.79

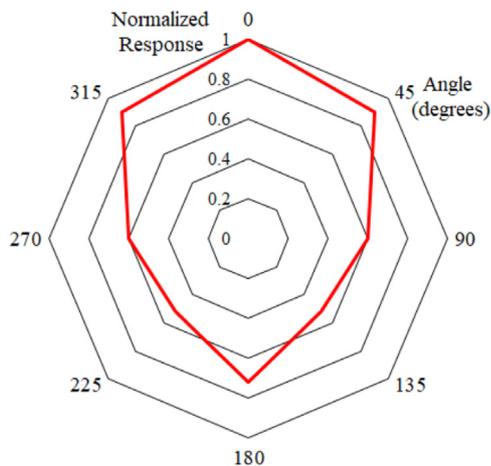


Fig. 9. Relative angular neutron sensitivity response measured using the Li Foil MWPC BRD with a 50 μ Ci bare-²⁵²Cf source.

Table 2
The raw data from the angular neutron sensitivity response measurements using a 50 μ Ci bare-²⁵²Cf source.

Detector orientation (degrees)	Instrument response (n/s)	Normalized response	Uncertainty
0	38.53	1.00	0.0077
45	34.70	0.90	0.0072
90	23.22	0.60	0.0054
135	19.96	0.52	0.0049
180	27.63	0.72	0.0061

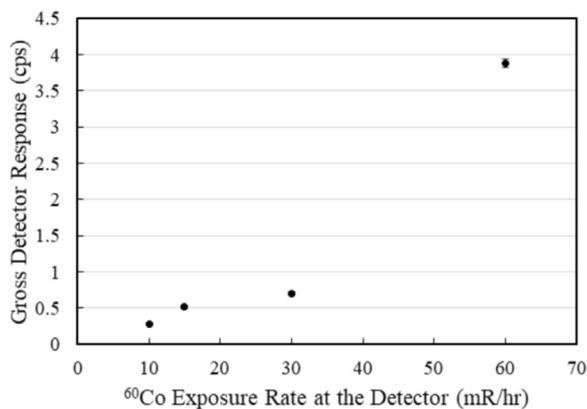


Fig. 10. The Li Foil MWPC BRD response as a function of ⁶⁰Co exposure rate at the detector.

Table 3
The Li Foil MWPC BRD response to gamma rays.

⁶⁰ Co source dose rate (mR/Hr)	Gross count rate (cps)	Gamma-ray rejection ratio
Background	0.1879 \pm 0.0026	–
10	0.278 \pm 0.015	6.90 $\times 10^{-9}$ $\pm 1.15 \times 10^{-9}$
15	0.5122 \pm 0.0239	6.85 $\times 10^{-8}$ $\pm 1.08 \times 10^{-8}$
30	0.7044 \pm 0.0280	5.40 $\times 10^{-8}$ $\pm 8.38 \times 10^{-9}$
60	3.8756 \pm 0.0654	1.95 $\times 10^{-7}$ $\pm 2.94 \times 10^{-8}$

source. The source-to-detector distance required to achieve a fluence rate of 1 n/cm²/sec was calculated to be 131 cm. The BRD was rotated through 180° in 45° increments, and 900-second measurements were collected. The normalized response over 360° is shown in Fig. 9, and the raw data is listed in Table 2. The shape of the resulting angular response is similar to the predicted results shown in Fig. 1, which further merits the preceding simulation efforts. The Li Foil MWPC BRD was also evaluated for gamma-ray response over a range of ⁶⁰Co exposures rates, including 10, 15, 30, and 60 mR/Hr and the detector response as a function of ⁶⁰Co exposure rate is shown in Fig. 10. The unshielded ⁶⁰Co source was placed at the necessary distance from the detector to achieve the desired exposure rate and a 900-second measurement was collected. The measured GRR ranged from 6.90 $\times 10^{-9} \pm 1.15 \times 10^{-9}$ to 1.95 $\times 10^{-7} \pm 2.94 \times 10^{-8}$, as listed in Table 3. The count rate measured from the 60 mR/Hr exposure rate was higher than what was expected for that exposure rate; the elevated count rate is suspected to be attributed to the malfunctioning of one of the Li Foil MWPCs that resulted in the device becoming noisy during that duration of the testing. The unfortunate result of using a Viton® gasket to seal the detector chambers inherently introduces high outgassing contaminants into the chambers where the lithium foils are positioned, and, thereby, increases the probably of lithium degradation and detector noise problems as a function of time. The Li Foil MWPC BRD was also evaluated for the gamma absolute rejection ratio in the presence of neutrons (GAARn), which resulted in a value of 1.00 as shown in Table 4. This measurement indicates that no statistical change of the Li Foil MWPC BRD absolute and intrinsic neutron detection efficiencies occurs in the presence of significant gamma-ray radiation.

5. Conclusions

The recent design and fabrication advancements to the Li Foil MWPC have increased the technology readiness level of the device, however, further advancements are continuing to be studied and implemented. Further work is currently underway to determine a reliable method for sealing the detector tanks and improve gas-fill methods. Laser welding is being explored, but is costly to implement. Other sealing methods, such as using pressure-fit lids with a low-outgassing epoxy, are also being explored. Li Foil MWPCs are still being fabricated using an inert atmosphere glove box. However, a dry room, similar to those used in the lithium battery industry, would be a substantial improvement in commercialization efforts by reducing the time required

Table 4
Neutron detection efficiencies as well as the measured GAARn result.

Radiation	Absolute efficiency (%)	Intrinsic efficiency (%)	GAARn
Neutron	1.86 $\times 10^{-2}$ $\pm 2.78 \times 10^{-3}$	5.85	1.00 \pm 0.0023
Neutron + Gamma	1.86 $\times 10^{-2}$ $\pm 2.79 \times 10^{-3}$	5.87	

to fabricate each Li Foil MWPC. This would allow for mass production of Li Foil MWPCs, whereas, in a glove box, only one or two detectors can be fabricated in a day due to space limitations of the glove box antechamber, which puts a size limit on what can be moved in and out of the glove box at one time. Additionally, the inherent reduction in dexterity when fabricating devices within a glove box substantially increases the time required to fabricate each device. The next phase of this project will explore these fabrication advancements to the Li Foil MWPC detector, in order to continue to refine and improve the infrastructure associated with bringing the technology to commercialization.

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