Glass-Panel ^{6}Li Neutron Detector

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Abstract— **We report on the development of a neutron detector utilizing solid enriched lithium, which has substantial neutron detection efficiency. The detector employs large, thin sheets of lithium in a gas-filled multi-wire proportional chamber (MWPC). Using low-cost design methods, readout electronics, and a small fraction of the already available enriched lithium available from Y-12/Oak Ridge National Laboratory, the amount of ^{3}He equivalent detection capability for nuclear non-proliferation activities can be greatly increased.**

^{3}He replacement technology; enriched lithium; thermal neutron detection

I. INTRODUCTION

For the past decade, there has been a heightened effort to minimize the risk of nuclear related terrorist attacks. A major focus of this effort has been to deploy radiation detectors within a larger security architecture. One important component of radiation detection is neutron detection, since neutron detectors can aid in the detection of fissile material such as plutonium. The standard method for neutron detection uses ^{3}He gas within a tube and signals read out by a central high voltage wire. The combination of high probability for the neutron/^{3}He interaction, full energy readout from the reaction, and the simple tube geometry makes the method a gold standard of neutron detection.

The limited supply of ^{3}He and the demand on its use for other activities such as medical imaging and low-temperature research has limited the number of neutron detectors that can be deployed [1]. Efforts are focusing on alternative technologies that can replace ^{3}He neutron detection. Benchmarks set in 2003 state that ^{3}He neutron detector replacements should be housed within the ^{3}He tube's envelope specified by already deployed radiation portal monitors, and that the replacement method should be as efficient as ^{3}He tubes within this geometry [2]. Development of alternative technologies has focused on these two requirements. Cost and scalability have played less of a role in the design of ^{3}He replacement technologies. This may limit the ability to expand to larger deployments of thermal neutron detection beyond the task of replacing ^{3}He units [2,3].

A recent Congressional report outlined what qualities of neutron detection technologies will be valuable within the upcoming decades, stressing the need for "future deployments, especially beyond the near term" where relaxing the "plug-and-play" needs of a ^{3}He replacement can be traded for a large reduction in price without sacrificing efficiency [1]. The goal of our research is to develop detectors whose low cost and larger surface areas allow them to surpass the benefits of more expensive detectors with higher intrinsic efficiencies.

Slow neutrons can be detected with high efficiency by a few special detector materials. Three nuclides are especially useful: ^{3}He, ^{6}Li, and ^{10}B. Helium is a rare gas with very little naturally occurring ^{3}He on earth. Boron is a solid metalloid element with a density of 2.34 g/cm^{3}. It does not react with air at room temperature. Natural boron consists of 20% ^{10}B and 80% ^{11}B. Lithium is a soft alkali metallic element with a density of 0.534 g/cm^{3}. It reacts with air at room temperature. Natural lithium consists of 7.5% ^{6}Li and 92.5% ^{7}Li. Each of ^{3}He, ^{6}Li, and ^{10}B has a large cross section, σ_{th}, for producing charged particle daughter products following absorption of thermal neutrons:

$$^{3}\mathrm{He} + n \rightarrow p + {}^{3}\mathrm{H} + 0.764\ \mathrm{MeV} \quad \sigma_{th} = 5372\ \mathrm{barns} \quad (1)$$
$$E_p = 0.573\ \mathrm{MeV},\ E_t = 0.191\ \mathrm{MeV}$$

$$^{6}\mathrm{Li} + n \rightarrow \alpha + {}^{3}\mathrm{H} + 4.786\ \mathrm{MeV} \quad \sigma_{th} = 940\ \mathrm{barns} \quad (2)$$
$$E_\alpha = 2.051\ \mathrm{MeV}\ (\mathrm{range} = 23.3\ \mu\mathrm{m})$$
$$E_t = 2.735\ \mathrm{MeV}\ (\mathrm{range} = 135\ \mu\mathrm{m})$$

$$^{10}\mathrm{B} + n \rightarrow \alpha + {}^{7}\mathrm{Li} + 2.79\ \mathrm{MeV} \quad \sigma_{th} = 234\ \mathrm{barns} \quad (3)$$
$$E_\alpha = 1.78\ \mathrm{MeV}\ (\mathrm{range} = 4.39\ \mu\mathrm{m})$$
$$E_{Li} = 1.01\ \mathrm{MeV}\ (\mathrm{range} = 2.04\ \mu\mathrm{m})$$

$$^{10}\mathrm{B} + n \rightarrow \alpha + {}^{7}\mathrm{Li}^{*} + 2.31\ \mathrm{MeV} \quad \sigma_{th} = 3603\ \mathrm{barns} \quad (4)$$
$$E_\alpha = 1.47\ \mathrm{MeV}\ (\mathrm{range} = 3.53\ \mu\mathrm{m})$$
$$E_{Li} = 0.84\ \mathrm{MeV}\ (\mathrm{range} = 1.82\ \mu\mathrm{m})$$
$$^{7}\mathrm{Li}^{*} \rightarrow {}^{7}\mathrm{Li} + 0.478\ \mathrm{MeV}\ \gamma\ (\mathrm{lifetime} \approx 100\ \mathrm{fs})$$

The ranges indicated above are in enriched ^{6}Li or ^{10}B, as appropriate. The mean free path for thermal neutron capture in enriched lithium (boron) is 229 mm (20 mm). The ratio of the triton (alpha) range to neutron capture mean free path in ^{6}Li is 59% (10%), while that of alpha (lithium) for ^{10}B is 18% (9%). It is thus much easier to detect triton products escaping from ^{6}Li than the neutron capture products from boron, giving lithium a large inherent advantage over boron as a neutron

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detector, as shown in detailed calculations by McGregor et al. [4].

Early investigations into the ^{6}Li MWPC neutron detectors were not successful at solving issues of vibrations of the ^{6}Li strips [5], or demonstrating that the detector design worked at the expected efficiency [4,6]. By partnering with industry leaders and using current techniques in readout electronics, we take advantage of years of advances that make the ^{6}Li foil MWPC more suitable than when it was first proposed in the late 1980s. Since then, the battery industry has motivated lithium companies to perfect the extrusion of thin lithium foils at thickness that are ideal for detector design, as well as develop methods of adhering lithium to surfaces. The double paned window industry manufactures low vibration, dry, argon filled environments between two panes of glass that are ideal for the gas readout portion of the detector. And low-noise charge sensitive preamplifiers allow a much better signal extraction at lower cost. Furthermore, the United States produced a total of 442.4 metric tons of enriched lithium from 1954 to 1963 for thermonuclear weapons, tritium production, and other purposes [7]. If only 5% of the material would be used for such detectors, 300k 1 m x 1 m (1-m^2) detectors can be made. Since two, 1-m^2 detectors surpass the capabilities of a ^{3}He tube radiation portal monitor, 150k ^{3}He efficient equivalent detectors could be deployed. Since there are about 1500 ^{3}He portal monitor stations used by the United States, glass-panel ^{6}Li detectors could increase portal thermal neutron detection capability for non-proliferation activities by two orders of magnitude. Lastly, the detectors can be made for a fraction of current ^{3}He replacement technology costs, further enabling a large amount of detectors to be deployed.

II. Detector Design

A. *Multiwire flat panel geometry*

The detector employs thin sheets or foils of lithium in a gas-filled multi-wire proportional chamber (MWPC). The lithium foil is sufficiently thin so that large particles created by reactions between incident neutrons and lithium nuclei escape the foil with sufficient energy to cause ionization of a detection gas (e.g., argon) in the chamber. Drift electrons from the ionizations are attracted to an array of wires in the chamber biased at a high voltage, and the resulting signals induced on the wires are detected and processed by electronic circuitry (Fig. 1).

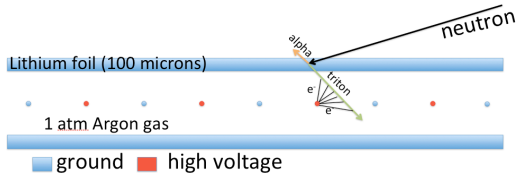

Figure 1. Schematic for the detector

This design, which is the so-called "double-outward" design, although simplistic, provides a relatively high level of intrinsic efficiency of 20% for thermal neutrons incident perpendicular to the surface of the lithium sheets [4]. The optimal thickness of the ^{6}Li layers are chosen for a given moderator housing and detector usage, and are within a range of 80 to 120 microns.

B. *Lithium procurement, handling and extrusion*

When in contact with humid air, lithium's surface develops a black coating consisting of LiOH, LiOH-H_2O, Li_2CO_3, and Li_3N [8]. This process can take seconds in humid air (60% relative humidity, or RH) to days in dry air (4% RH) and indefinitely within environments below 2% RH. It is theorized that this permanent resistance to tarnish in environments below 2% RH is due to a transparent film of reaction products which form a protective barrier on the lithium surface [9]. It is not difficult to work with lithium in dry glove boxes or in rooms with relative humidity restricted to less than 2% RH. Furthermore, double-paned, argon filled window environments which retain a 0.01% RH (-70°C dew point) moisture level for 10 or more years [10] are appropriate housing for exposed lithium surfaces.

We have obtained 5.5 kg of ^{6}Li from Y-12/Oak Ridge National Laboratory for the research and development of the detector, as well as for the initial production models that will be made in the upcoming years (Fig. 2).

Figure 2. 440 g of lithium "chunks" purchased from Y-12/Oak Ridge National Laboratory, which were received in mineral oil and had a thin (100 micron) blackened layer. The chunks were cut open for rolling experiments. The open surfaces reveal the silvery-white untarnished lithium material.

The Y-12/Oak Ridge lithium is enriched to 95.58 atomic percent with less than 100ppb of Uranium and Thorium contaminants. Lithium can either be rolled or extruded to reduce its thickness to the desired range of 100 microns. The lithium battery industry has developed best practices for lithium extrusion to thicknesses used in the detector design. Their methods maximize the strength and durability of the produced foils, which is advantageous during battery manufacturing. We are working with Lithium Innovations, LLC of Toledo Ohio to develop best practices for enriched lithium processing. After creating an ingot from the material, Lithium Innovations extruded 300 g of 1" wide foil, where they tested foil production from 90 to 125 microns with 10 micron variances (Fig. 3). This produced 300 meters of foil, enough ^{6}Li for 3 1-m^2 detectors, with a follow-up of an

additional 5 kg of enriched lithium, which will be enough to manufacture 50 1-m^2 detectors.

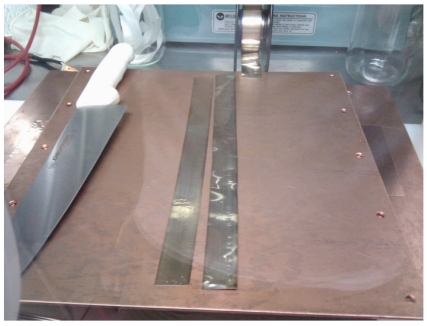

Figure 3. Process of placing lithium strips onto copper pieces within the prototype chamber. The lithium is pulled using tweezers from the spool of lithium, then cut with a kitchen knife at the appropriate length.

C. *Detector materials and sealing*

From literature reviews of the common reactions of lithium with other materials [11] as well as our own tests (see Fig. 4), we have determined what materials can be used within the detector design. These materials are aluminum, copper, stainless steel, glass, butyl rubber, poly-isobutylene (PIB), and low outgassing epoxy. Several materials that create reactions that destroy the lithium are acrylic, silicone, and circuit board material.

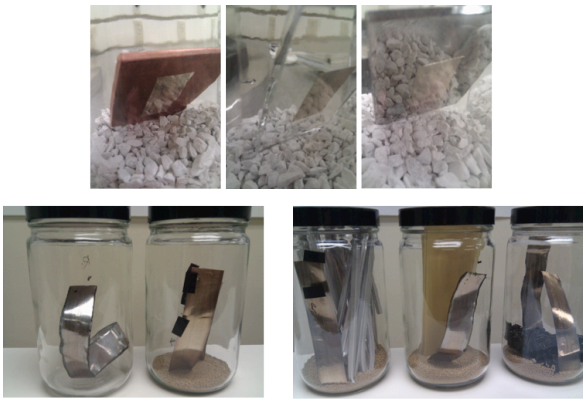

Figure 4. (Upper) ^{6}Li strips on copper, glass, and stainless steel. Each piece is in a seal lid jar on a bed of calcium sulfate ($CaSO_4$). The ^{6}Li has been in this state on the materials for two years. (Lower left) ^{6}Li placed in jar with and without 30 mL of molecular sieve desiccant for 1 year – there is no degradation of the lithium with the desiccant. (Lower right) ^{6}Li with desiccant with aluminum, circuit board material, and poly-isobutylene (PIB). Only the ^{6}Li with the circuit board material shows degradation.

III. Prototype Chamber

A. *Design*

We have built a prototype detector to test the detector concept (Fig. 5). The prototype has a 1 cm gas gap, and 5 mm pitch between the field shaping and anode wires that are held at positive high voltage. The active region has dimensions 20 cm by 30 cm. Copper plates support lithium foils as the cathode surfaces. Wires are attached under tension to a circuit board that maintains a 5 mm gap between the wires and one cathode plane. Spacers provide a gap of 5 mm between the wires and the other cathode plane.

One inch thick slabs of high density polyethylene (HDPE) are attached to the copper plates to stiffen the assembly and to moderate fission neutrons to thermal energies. The preamplifiers and summing amplifiers were designed and built in the Boston University (BU) Electronics Design Facility (EDF). The preamplifiers have gain 1 mV/fC, and input noise of 2000 electrons. We found that noise within and outside a Faraday cage were not significantly different.

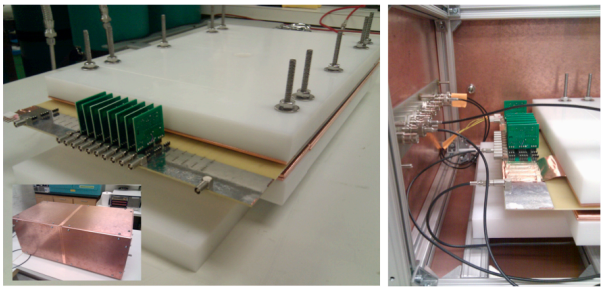

Figure 5. Prototype detector with high-density polyethylene (HDPE) top and bottom supporting the copper cathodes. 8 preamplifiers are installed to read out the signals from 8 high voltage wires from the multi-wire proportional chamber (MWPC). The placement within and outside of the copper faraday cage box tested the intrinsic radio frequency pickup of the detector design.

We placed enriched lithium foils on the copper cathode plates from a spool of one inch wide, 120 mm thick, 23 m long lithium extruded by Lithium Innovations. Due to lithium's greater affinity to adhere to copper than HDPE, we used a HDPE rolling pin to flatten the lithium onto the copper plates (Fig. 6). Conductive copper tape connects the copper/lithium to the ground plane of the circuit board. Two 30 A-hr batteries provide negative and positive low voltage to the preamplifiers and the summing amplifier, and a voltage supply outside the glove box provides high voltage for the anode wires. The gamma ray background is below 110 keV for the prototype, well below that of comparable solid-state diode detectors.

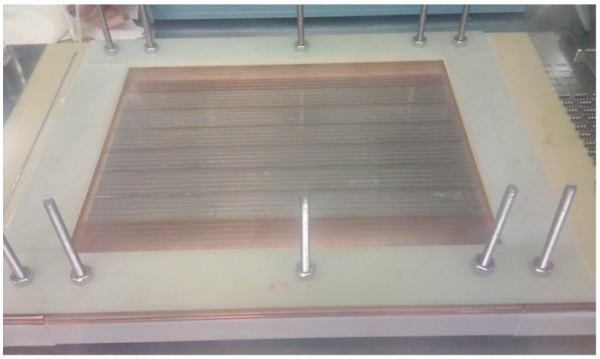

Figure 6. An HDPE rolling pin was used to flatten the lithium onto the bottom and top (not shown) copper plates before the wire frame is put in place.

We calibrated the prototype with a ^{241}Am source that illuminates a 1 mm diameter hole in the top cathode plate with 4.8 MeV alpha particles. The alphas deposit 1 MeV of energy in the 1 cm Ar gas at a pressure of 1 atm. We use 950 volts on the anode wires, providing a gas gain of 100 (Fig. 7).

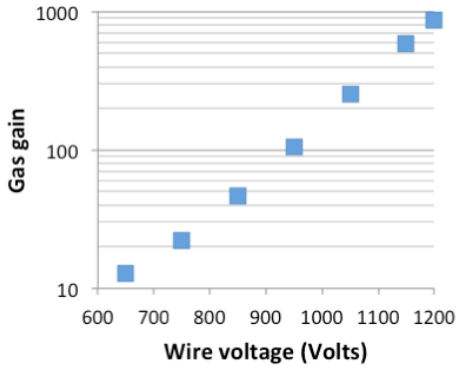

Figure 7. Gas gain as a function of voltage applied to the readout wires within the detector.

We have measured the neutron detection efficiency with a 2 ng ^{252}Cf source (1.08 μCi). The source is placed below the glove box, three inches from the MWPC (Fig. 8), with HDPE placed between the source and the MWPC. The observed counting rate above 110 keV increases to 16 Hz with the source in place. Fig. 9 shows the spectrum for the lithium MWPC. Also shown are results from a Geant4 simulation. The peak at 300 keV is due to the large number of tritons that escape with a large fraction of their initial energy. They do not stop in the gas due to their large energy, so the energy deposited is the product of dE/dx and the gas thickness. The simulations imply the detection efficiency of the lithium to be 25%. This is consistent with predictions by McGregor et al [4].

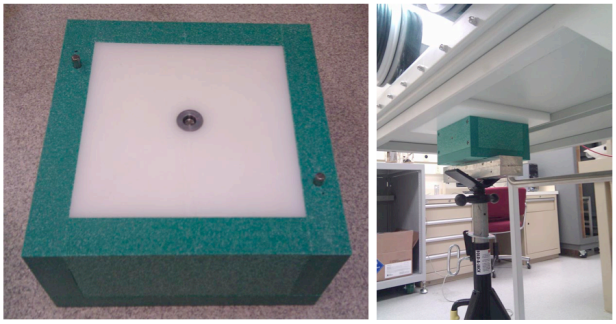

Figure 8. ^{252}Cf source in HDPE housing. The green polyethylene is doped with boron. The source is supported by a jack below the glove box to expose the MWPC with neutrons at a distance of three inches. Three inches of white HDPE separate the source from the MWPC.

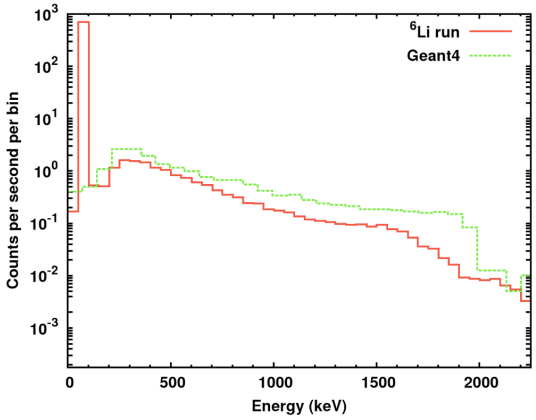

Figure 9. Spectrum from exposure of enriched lithium MWPC to ^{252}Cf neutron source from data and from a Geant4 simulation. The total counts between 110 keV and 2500 keV were 29125 for 1841 seconds for the data, and 34916 for the same time for the simulation.

We have also tested a MWPC with enriched boron carbide having the same dimensions as the lithium MWPC (i.e. 20 cm by 30 cm). The thickness of the boron carbide layer was 2.75 mm. The results are shown in Fig. 10. The count rate is about 6 Hz, showing the increased efficiency of lithium compared to boron carbide.

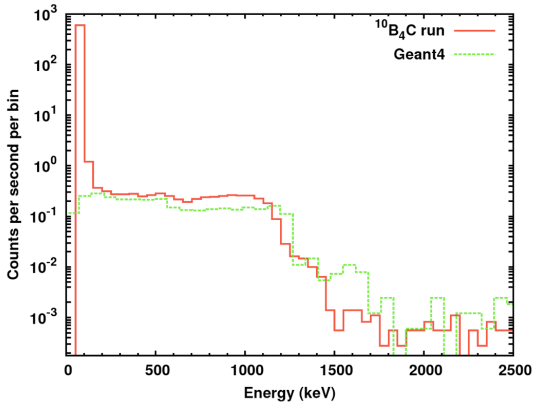

Figure 10. Spectrum from exposure of enriched boron carbide MWPC to ^{252}Cf neutron source from data and from a Geant4 simulation. The dimensions were the same as for the lithium detector results of Figure 9. The total counts between 110 keV and 2500 keV were 22820 for 3608 seconds for the data, and 24281 for the same time for the simulation.

IV. Full Size Detector Design

A. B_4C field-testable detector

In order to test several detector features such as the high-tension wires, sealed internal gas environment, preamplifiers, summing amplifiers, and readout electronics, we built two small MWPCs using enriched boron carbide on stainless steel plates in the place of the ^{6}Li on glass design (Fig. 11). The

detectors have a 4"x4" active region utilizing 2.75 micron thick enriched boron carbide. The argon gas environment is sealed using epoxy, and gas valves allow the flushing of the gas environment.

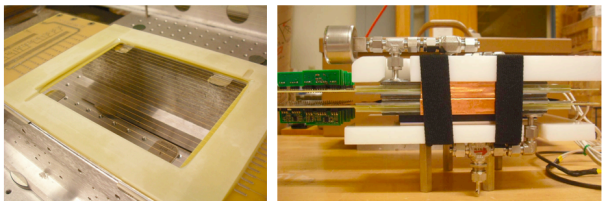

Figure 11. Enriched boron carbide (B_4C) detectors. (Left) An intermediate stage where the wires and non-conductive G-10 spacer are being epoxied to the stainless steel frame. (Right) Two final B_4C detectors stacked on top of one another with gas fixtures and HDPE above and below for neutron moderation. The preamplifiers are at the ends of the wires that feed through the end of the gas volume of the detector.

B. Lithium window development

The double paned window industry has developed methods to create sealed argon gas environments between two panes of glass [10]. A sealed argon environment keeps moisture from in-between the window panes so that fogging does not occur, and provides a gas compound that reduces heat loss through windows. The standard quality of these seals keeps a dew point of below -70°C, and allows 1% argon loss per year. We determined that the double paned window sealing practices using an aluminum channel for molecular sieve desiccant with a gas tight seal created with low outgassing epoxy provides a proper environment for the lithium (Fig. 12).

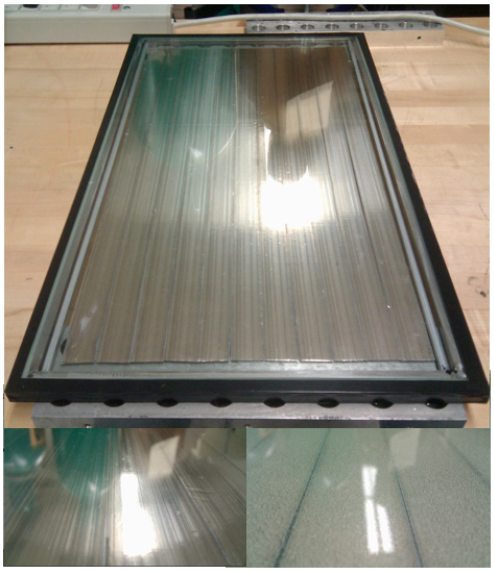

Figure 12. (Top) A 23"x13" Lithium double paned glass panel which employs sealing and dessicant channels developed by the window industry. A sheet of 1" lithium strips are epoxied to one of the internal window surfaces. (Bottom) left and right show a zoom in of the smooth front and epoxied back surface of the lithium. The lithium can be adhered to glass surfaces with flatness tolerances that exceed those needed for the proportional chamber environment. These systems show no sign of lithium degradation for 1 year.

C. Microphonics

From the design of the first generation detector, we are aware of microphonics issues that may be involved with wire and ^{6}Li vibrations. Microphonics issues have been a challenge for researchers of this method in the past [5]. In recent years there have been advances in the controlled tensioning of wires using wire-winding machines. High-tension wires are less susceptible to microphonics due to their higher vibrational modes. We have acquired the services of Brookhaven National Laboratory to provide 450 gram high tensioned wires using the wire-winding method. 50 micron diameter gold plated tungsten-rhenium wires are stretched and stored on wire transfer frames before they are epoxied into individual detectors. The wire winding production is easily expanded to production environments, as shown during the manufacture of flat panel detectors for the Large Hadron Collider (LHC).

D. Next generation full size detector

Using the techniques outlined above, we plan to design and build a full scale, square meter detector within the next two years. Fig. 13 shows a rendered design of such a chamber.

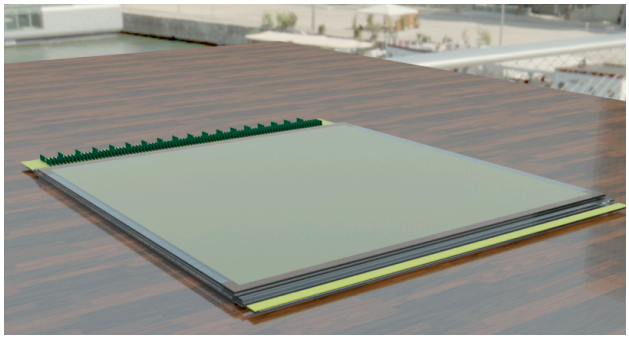

Figure 13. Rendered design image of 1-m^2 flat panel, sealed, double-paned window, lithium MWPC chamber.

We use Geant4 to find the efficiency and analyze the properties of the 1-m^2 flat panel, sealed, double-paned window, lithium MWPC chamber. The detector test as defined in [2] consists of a ^{252}Cf source 2 m away from the center of a 1-m^2 detector, both of which are 1.5 m above grade (Fig. 14). The neutrons from the spontaneous fission of ^{252}Cf radiate isotropically and when an alpha or triton particle is found in the argon sensitive readout section, a "count" is recorded. With an energy threshold of 100 keV, the count rate is 2.61 cps/ng, and with an energy threshold of 300 keV the count rate is 2.33 cps/ng. These values are below the requirements for ^{3}He replacements [2], however two of these detectors side by side would double the surface area and therefore have an efficiency of 4.4 cps/ng in this testing geometry.

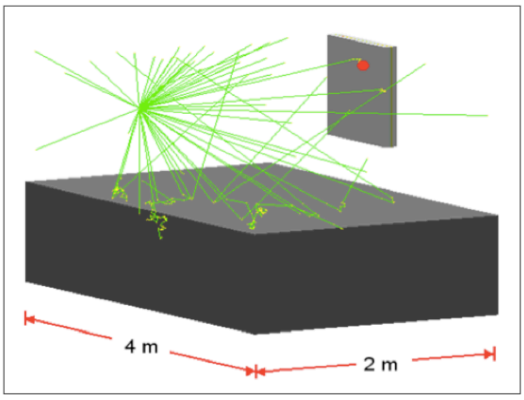

Figure 14. Geometry for the simulated test of the neutron detector. A ^{252}Cf emits neutrons isotropically (green rays) 2 m away from the center of a 1-m^2 lithium panel detector, both of which are 1.5 m above a concrete platform.

Table 1 shows the material costs for a 1-m^2 detector.

TABLE I. DETECTOR MATERIAL COSTS

Description	Cost
75 grams of ^{6}Li at $2.45 per gram[a]	$200
Extrusion of ^{6}Li	$100[b]
Electronics	$300
Metal/glass/dessicant/epoxy structure	$200
Wire material and wire stretching	$200
Total	**$1000**[c]

a. Current price from Y-12/Oak Ridge National Laboratory

b. Estimate based on market costs of primary lithium batteries

c. Does not include labor (~10 man-hours) or facilities cost

V. CONCLUSION

Since the 1980s, the battery industry has perfected the extrusion of thin lithium foils at thickness that are ideal for lithium metal neutron detectors. The double paned window industry has motivated the manufacturing of low vibration, dry, argon filled environments between two panes of glass that are ideal for the gas readout portion of the detector. And low-noise charge sensitive preamplifiers allow better signal extraction at lower cost. Furthermore, there is a stockpile of enriched lithium metal ready to be used for such a detection method, removing the need for high-cost enrichment and sputtering procedures. The combination of new technologies, simplistic MWPC design, and readily available detection material gives an effective, cost-efficient and scalable solution to thermal neutron detection.

ACKNOWLEDGMENTS

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REFERENCES

[1] D.A. Shea and D. Morgan, "The Helium-3 Shortage: Supply, Demand, and Options for Congress," Specialist in Science and Technology Policy, Congressional Research Service, December 2010.

[2] R.M. Van Ginhoven, R.T. Kouzes, and D.L. Stephens, "Alternative Neutron Detector Technologies for Homeland Security," Pacific Northwest National Laboratory Report, June 2009.

[3] R.T. Kouzes et al., "Alternatives to ^{3}He for Neutron Detection for Homeland Security, " Pacific Northwest National Laboratory Report.

[4] D. S. McGregor, M. D. Hamming, Y. H. Yang, H. K. Gersh, and R. T. Klann, "Design considerations for thin film coated semiconductor thermal neutron detectors-I: basics regarding alpha particle emitting neytron reactive films," Nuclear Instruments and Methods in Physics Research A, vol. 500, pp. 272-308, 2003.

[5] "An investigation of parallel plate neutron detector concept," Final report IRT 8169-004, IRT Corportation, 1978.

[6] R. Engles, G. Kemmerling, H. Noldgen, and J Schelten, "Thermal Neutron Detection with 6-Li Converters," IEEE Nuclear Science Symposium Conference Record, 2007.

[7] Declassification of the Quantity of Enriched Lithium Produced at the Y-12 Plant in Oak Ridge, Tennessee https://www.osti.gov/opennet/forms.jsp?formurl=document/press/pc23.html#ZZ6

[8] R. Rhein, "Lithium Combustion: A Review," Naval Weapons Center Report, December 1990.

[9] V.V. Rachev, B.K. Maslennikov, and A.A. Lbov, "Behavior of the Surface of Metallic Kithium in Air and Argon of Low Humidity," Zh. Prik. Khim., 1962, vol. 35, p.189.

[10] G.R. Torok and A.L. Major, "Predicting Time to Fogging of Insulating Glass Units," Canada Mortgage and Housing Corporation Research Report, August 2005

[11] D.W. Jeppson, J.L. Ballif, W.W. Yuan, and B.E. Chou, "Lithium Literature Review: Lithium's Properties And Interactions," Hanford Engineering Development Laboratory Report, April 1978.