1 Reliability of Monte Carlo simulations in modeling neutron yields from a shielded fission source

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Abstract

Using the combination of a neutron-sensitive ^6Li glass scintillator detector with a neutron-insensitive ^7Li glass scintillator detector, we are able to make an accurate measurement of the capture rate of fission neutrons on ^6Li . We used this detector with a ^{252}Cf neutron source to measure the effects of both non-borated polyethylene and 5% borated polyethylene shielding on detection rates over a range of shielding thicknesses. Both of these measurements were compared with MCNP calculations to determine how well the calculations reproduced the measurements. When the source is highly shielded, the number of interactions experienced by each neutron prior to arriving at the detector is large, so it is important to compare Monte Carlo modeling with actual experimental measurements. MCNP reproduces the data fairly well, but it does generally underestimate detector efficiency both with and without polyethylene shielding. For non-borated polyethylene it underestimates the measured value by an average of 8%. This increases to an average of 11% for borated polyethylene.

keywords: Neutron detection; Portal monitor; Homeland security; Room return; Monte Carlo modeling; MCNP

1. Introduction and Motivation

Currently, radiation portal monitoring (RPM) systems are used to help prevent the spread of special nuclear materials. Neutron detectors are an integral part of these systems since they can detect the presence of nuclei such as ^{239}Pu , which have high spontaneous fission rates. It is important that these detectors are sensitive not only to bare fission sources, but also to shielded sources because illicitly imported material will likely be shielded to avoid detection. Kouzes, *et al.* [1] performed extensive MCNP (Monte Carlo Neutral Particle) [2] simulations of the response of moderated ^3He detectors to fission sources embedded in a variety of shielding materials. Rees and Czirr [3] extended these simulations to include several moderating strategies for ^3He tubes detecting neutrons emitted from fission sources shielded with polyethylene. Tomanin, Peerani, and Janssens-Maenhout [4] along with Gilbert, *et al.* [5] have done additional computational studies to optimize ^3He detectors for national security applications. When MCNP simulations are used to model shielded sources, they often require the modeling of multiple neutron interactions. Small discrepancies in cross sections or modeling techniques could possibly lead to significant deviations from experimental data. An experimental measurement of neutron flux from shielded sources is important as a check of the validity of such simulations.

The results of such experiments are dependent on the shielding and the detectors that are used. Polyethylene is an inexpensive, efficient, and readily available neutron moderator and is used in many neutron shielding applications because of its high hydrogen content. High hydrogen content is desirable because neutron-hydrogen collisions cause neutrons to lose an average of about half of their energy per collision. To improve the absorption of neutrons in the polyethylene, boron, which has a very high neutron capture cross section, can be added. Borated polyethylene has been found to be a considerably better shielding material [6] than non-borated polyethylene. We constructed a moderated lithium-glass, Fig. 1, that is sensitive to both high and low energy fission neutrons. Using this detector, we measured the effects of shielding a ^{252}Cf fission source with both non-borated polyethylene and borated polyethylene.

[Figure 1]

2. Experimental Methods

2.1 Detectors

The choice of detector in this measurement is important as the results of both the experiment and the simulation are very detector dependent. The following criteria were the most important:

- 1) Since the energy of neutrons from shielded sources can vary from thermal to over 10 MeV, the detector should be sensitive to a wide range of neutron energies.
- 2) The signals produced by low-energy neutrons should not be small enough to be confused with noise.
- 3) The detector should produce one pulse for every neutron that MCNP would count as detected. If a simulation has to calculate light output and light transmission to determine if a pulse reaches a threshold value, it is much less likely to be reliable.
- 4) The detector must be able to distinguish neutrons from gammas either event by event or in the aggregate.

A variety of detectors could meet these criteria. A ^3He detector placed in a box with 5 cm-thick polyethylene walls, a typical moderator [1], does not meet the first criterion as it has little sensitivity to thermal neutrons [3]. Organic scintillators produce energy-dependent signals, and where neutron-gamma discrimination is possible, the discrimination is not good when pulse heights are small. Our choice was to build detectors with lithium glass and lucite moderators. ^6Li has a large cross section for neutron capture and has been used in a number of neutron detector applications including glass fibers [7], liquid scintillators [8], plastic scintillators [9], and lithium glass [10].

Two neutron detectors were used in conjunction with each other for this study. The first consisted of two sheets of ^6Li -doped glass scintillator (Applied Scintillation Technologies GS-20, lithium is 95% ^6Li), each 1 mm thick and 12.7 cm in diameter. The first piece of glass was placed at the front of the detector followed by a 2.5 cm-thick lucite moderator. Lucite was chosen because it has a reasonably large hydrogen content, it is optically clear, it is easily machined, and it is available in convenient sizes. The second sheet of glass scintillator was placed on top of the lucite and then an additional 7.6 cm-thick lucite moderator was added. The glass and lucite all have 12.7 cm diameters. Finally, we place a 12.7-cm diameter Adit Model B133D01S photomultiplier tube on top of the second lucite moderator, as shown in Fig. 1. The pulses of the photomultiplier tube are recorded by a signal digitizer so we can restrict the analysis to pulses with areas consistent with neutrons.

The second detector was identical to the first, except that the ^6Li glass was replaced by ^7Li glass (Applied Scintillation Technologies GS-30, lithium is 99.99% ^7Li). The sheets of GS-20 and GS-30 are identical except for the isotopic composition of the lithium. The ^7Li detector is used to remove ambient and source-related gamma backgrounds from the neutron signal, as described below. This technique was used because the ^6Li detector is incapable of clean event-by-event neutron-gamma discrimination.

The ^6Li in the glass scintillator has a large capture cross section for lower-energy neutrons. The neutron capture and subsequent fission process is as follows:

The alpha particle has a range of 6.2 μm in the lithium glass and the triton has a range of 35.2 μm . Therefore, both particles will generally deposit all of their energy in the 1 mm-thick piece of glass. The intensity of the light pulse produced by the scintillator in the glass is constant for all detected neutrons. This is because the neutron kinetic energy is typically very small for capture events so that the final-state charged particles always have essentially the same energies. There is a little variation in signal size, but this is more a matter of optical considerations. This can be seen in Fig. 2, a histogram of the peak areas for both ^6Li and ^7Li detectors, where the neutron peak from ^6Li is seen clearly on the right.

[Figure 2]

Two sheets of glass scintillator are used because the incident neutrons can vary greatly in energy. Thermal neutrons will almost all be absorbed by the first sheet of glass. Most neutrons with energies of roughly 100 keV or higher pass into the lucite where they are moderated. Once moderated, these neutrons can be captured in either piece of glass. Monte Carlo simulations [3] show that the neutrons usually lose most of their energy within two or three collisions and then undergo a random walk within the moderating material. This effectively produces a “neutron cloud” within the moderator. The depth of the cloud depends on the energy of the incident neutrons. We place the second sheet of glass scintillator at a depth that is optimized for 250 keV neutrons. This energy is lower than the peak of the energy distribution for a bare source, but is a good choice for neutrons from a shielded source. An MCNP simulation of the relative efficiency of each sheet of glass for absorbing neutrons is shown in Fig. 3. The shielding thickness reported throughout the paper is the thickness of the shielding on either side of the source, so the total thickness is twice that amount. The neutrons from the unshielded source have sufficiently high energy that 70% of all neutrons captured in lithium glass are captured in the second glass sheet; however, as shielding surrounding the source increases and the energy of the neutrons leaving this shielding decreases, nearly all of the neutron capture occurs in the first sheet of lithium glass. This allows for high sensitivity over a broader range of neutron energies. A Monte Carlo simulation suggests that the efficiency for the simple detector shown in Fig. 1 (with no can) to normally incident neutrons is 66% for 1-eV neutrons, 17% for 100-keV neutrons, 7.8% for 1-MeV neutrons, and 1.1% for 10-MeV neutrons.

While these detectors work well in the specific application for which they were designed, they are not suitable for portal monitoring [11] as they cannot accurately determine if an individual event is from a gamma or a neutron. Furthermore, the detection strategy required acquisition times of about 30 minutes to obtain good enough statistics to determine an accurate neutron count, a time that is too long for practical national security applications.

[Figure 3]

2.1.1 Gamma ray backgrounds from the source and from the ambient environment.

As can be seen in Fig. 2, the neutron signal from the ^6Li detector is not completely clean. The counts to the left of the neutron peak come from gamma rays. There are two sources of gammas, gammas from the californium source (associated with both fission and from ^{248}Cm), and ambient background gammas. Additionally, there are a small number of cosmic ray neutrons that are detected and lie in the neutron peak as well.

Gamma rays can undergo Compton scattering in the glass and transfer energy to electrons which in turn excite the scintillator and create light pulses. The maximum range of Compton electrons from 1-MeV gammas in the glass is 1.6 mm. In most events only a fraction of the electron energy is deposited within the glass itself, so gamma pulses tend to be smaller in

amplitude than neutron pulses, as can be seen in Fig. 2. However, some photons can produce pulses that are similar in size to the neutron pulses. Although this gamma background is typically not large, we must account for it. [12]

The ^7Li detector was used to measure the gamma background. The ^7Li glass scintillator has an identical gamma sensitivity to the ^6Li detector, but it is insensitive to neutrons since it lacks the high neutron absorption cross section of ^6Li . This is then used in conjunction with the first detector to allow the subtraction of gamma events.

The gains of the two detectors were matched by using a ^{60}Co source and adjusting the operating voltages of the photomultiplier tubes. Every set of shielding data consisted of four sets of measurements. Two background runs (no source) were taken, first using the ^6Li detector and then the ^7Li detector. Then, two runs with the ^{252}Cf fission source were also taken, again with the ^6Li then the ^7Li detector. The signal from the ^7Li with the ^{252}Cf source is also shown in Fig. 2. The total number of source neutrons detected, T , was then calculated as follows:

$$T = [^6\text{Li}(\text{Cf}) - ^7\text{Li}(\text{Cf})] - [^6\text{Li}(\text{Bkg}) - ^7\text{Li}(\text{Bkg})] \quad \text{Eq.2}$$

The $^6\text{Li}(\text{Cf})$ term includes source neutrons, ambient background neutrons, source gammas, and ambient background gammas. The $^7\text{Li}(\text{Cf})$ term includes only source gammas and ambient background gammas. Similarly, the $^6\text{Li}(\text{Bkg})$ term includes ambient neutrons and gammas, but $^7\text{Li}(\text{Bkg})$ includes only ambient gammas. What remains is only the source neutrons. The number of counts in the neutron region that is subtracted as background ranged from 3.4% (3.0 cm of non-borated polyethylene shielding) to 25% (20.3 cm of borated polyethylene shielding) of the total counts in the region.

The two-detector design creates a high level of confidence in our measurement of the total number of source neutrons detected, which allows us to calculate accurately the efficiency of the detector.

2.2 Source and shielding

Our neutron source was a 88.96 μCi (original activity) calibrated ^{252}Cf source. The activity has a 3.2% uncertainty at the 99% confidence level. We calculated its activity at the time of the experiments by using a half-life of 2.645 years. Neutron flux on the detector was calculated using a branching ration for fission of 3.092%, and a neutron multiplicity of 3.7675 neutrons per fission.

30.5 cm $\times$ 30.5 cm $\times$ 2.5 cm slabs of borated polyethylene and 30.5 cm $\times$ 30.5 cm $\times$ 3.0 cm slabs of non-borated polyethylene were used as shielding. The source was placed in a small slot in the center of two slabs of shielding. Data were taken with the bare source and with two, four, six, eight, ten, twelve, and sixteen slabs of shielding *equally divided above and below the source*.

2.3 Experimental Setup

Another difficulty with making accurate neutron measurements is that of "room return" caused by walls, floors, etc., that readily reflect neutrons and cause overestimates of detector efficiency. To minimize this effect, we placed our detector, shielding, and source on top of a 6.1 m scissor lift. This removed our experiment from all walls, floors, and other large objects that could backscatter neutrons into the detector.

On the scissor lift, the detector was placed facing downward on a thin steel sheet 1.2 m above the floor of the lift. The source was elevated 48 cm above the lift floor and 55 cm below the detector.

MCNP was used to model each of these configurations. The chemical makeup of the borated polyethylene was provided by the distributor and verified using x-ray photoelectron spectroscopy. By weight percentage, the borated polyethylene used was 11.60% hydrogen, 62.20% carbon, 22.00% oxygen, and 5% boron.

3. Results/Data

3.1 Non-borated polyethylene

We describe our results in terms of "practical intrinsic efficiency," defined as the ratio of detected neutrons to neutrons emitted from the source into the solid angle subtended by the detector. Thus the practical intrinsic efficiency is reduced by

absorption of neutrons in the shielding. In this paper, the practical intrinsic efficiency is integrated over the entire energy spectrum of fission neutrons from ^{252}Cf . The detector has a maximum measured practical intrinsic efficiency of 53.5% with 3 cm of non-borated polyethylene shielding both above and below the source. The decrease in the neutron detection rate with larger amounts of shielding occurs solely because of the large number of neutrons that are absorbed in the shielding. With more shielding, the energy distribution shifts to lower energies, while the number of neutrons absorbed in the shielding increases. Although the lower energy neutrons emerging from the additional shielding are detected more readily, the practical intrinsic efficiency decreases because the neutron absorption in the shielding dominates the process. Figure 4 shows an MCNP-PoliMi [13] simulation of the energy distribution of neutrons leaving the front surface of the polyethylene. Note that the shape of the distribution varies little as the shielding increases from 2.5 cm to 17.5 cm on each side of the source. Results for borated polyethylene are very similar, but the overall absorption is larger.

The lowest measured practical intrinsic efficiency with non-borated polyethylene shielding was taken with 24 cm of shielding on each side of the source. At this level of shielding, the practical intrinsic efficiency was measured to be 1.4%. Although the efficiency has dropped off considerably, the neutron counting rate is over 100 times the measured neutron background rate.

[Figure 4]

The MCNP simulations showed a general tendency to underestimate the measured efficiency, as seen in Fig. 5. In this figure the solid line represents the MCNP simulation, the circles are the experimental measurements, and the dashed curve is an MCNP-guided fit to the experimental data. Statistical uncertainties in the experimental measurements are much smaller than the symbols. The bare source comparison showed an MCNP underestimation of 9%. With only one slab of shielding this underestimation dropped to 2% and then gradually increased with increased shielding until it reached 21% at 24 cm of shielding.

[Figure 5]

Although the MCNP prediction is fairly close to the bare source measurement, the fact that it underestimates the experimental efficiencies suggests either systematic errors in the experiment or discrepancies in the modeling of the detector itself. As we add one layer of shielding, the difference between experiment and MCNP decreases. It is possible that MCNP underestimates the effects of small amounts of shielding; however, it may also mean that MCNP is more accurate in calculating the detector response for the softer energy spectrum of the moderated neutrons. As the shielding increases, the neutron energy spectrum does not change very dramatically, as seen in Fig. 4, but the MCNP simulation becomes increasingly smaller compared to experiment. It is likely that this results from MCNP overestimating neutron absorption in the polyethylene as the number of neutron interactions increases in the moderating material.

3.2 Borated Polyethylene

With 5% borated polyethylene, the neutron detection rate was about a factor of two less than that obtained with non-borated polyethylene shielding in the same configuration. This is illustrated in Fig. 6 where the practical intrinsic efficiency of the detector is shown for both non-borated and borated polyethylene shielding. The dashed curve is the ratio of the MCNP-guided fits to the data for borated to non-borated polyethylene. The ratio is constrained to be 1.0 with no shielding but quickly becomes about 0.5 and remains near that value through the entire range of shielding thicknesses.

[Figure 6]

With borated polyethylene shielding, the detector's practical intrinsic efficiency reached a maximum at 27% with 2.5 cm of shielding. It then followed a similar curve to non-borated polyethylene ending at 1.3%. With maximum shielding, the neutron detection rate was still about 30 times the background neutron rate. As seen in Fig. 7, MCNP once again showed a tendency to underestimate the measured efficiency. As in Fig. 5, the solid line represents the MCNP simulation, the circles are the experimental measurements, and the dashed curve is an MCNP-guided fit to the experimental data.

[Figure 7]

With 2.5 cm of borated shielding, MCNP underestimates the measured efficiency by 4% and at 5.1 cm of shielding it underestimates the experiment by 8%. This number rapidly increases to about 20% for 7.6 cm to 15.2 cm of shielding, but overestimates the data by 19% at 20.3 cm of shielding.

The energy spectrum of neutrons from the borated polyethylene is similar to that for non-borated polyethylene. The difference is primarily due to neutron absorption by the boron. So the comparison to the non-borated polyethylene provides a test of MCNP's calculation of the neutron energy spectrum combined with its boron absorption cross sections. The agreement with experiment is a little poorer here than with non-borated polyethylene, with the discrepancy increasing with shielding thickness except at the largest thickness. If the boron cross sections are accurate, then it would appear that MCNP also predicts too soft a spectrum for multiple neutron interactions in polyethylene so that boron absorption is overpredicted. However, the last data point does not follow the same systematics, so this interpretation is not certain.

3.3 Systematic Errors

There are several potential sources of systematic errors in these measurement. We will consider each of these in turn.

Since ${}^6\text{Li}$ and ${}^{10}\text{B}$ have very large neutron capture cross sections, we need to consider how significantly the results depend on the amount of lithium and boron that are included in the simulations. We corroborated the manufacturer's specifications for the composition of the borated polyethylene by measuring elemental concentrations with x-ray photoelectron spectroscopy. But because there can be some inhomogeneity in the boron distribution, we performed MCNP simulations of neutron absorption in boron for 2.5 cm of borated polyethylene shielding above and below the source with boron concentrations ranging from 0% to 10% boron (natural isotopic abundance) by weight. These data are shown in Fig. 8. The boron is included as a borate (BO_3) and the total density was fixed to match the experimental value. The decrease in absorption for higher amounts of boron is the consequence of reduced hydrogen, and hence less neutron moderation. If we do not decrease the hydrogen concentration as we increase the boron concentration, the curve remains flat from about 2% boron to 10% boron. This appears to be the result of even small concentrations of boron being adequate to absorb neutrons once they are moderated sufficiently for the neutron absorption cross section to be large. A change in boron concentration of 20% results in a change of only 2% in the MCNP efficiency calculation.

[Figure 8]

We used the manufacturer's specifications for the composition of the ${}^6\text{Li}$ glass. To see how important the lithium concentration is to the Monte Carlo results, we performed MCNP simulations with varying amounts of lithium in the glass as well. An increase of 10% in ${}^6\text{Li}$ concentration produced a change of 2.5% in the Monte Carlo efficiency, so there is little sensitivity to lithium concentration either.

The subtraction of all background contributions must be done carefully. Ambient neutrons ranged from 0.2% to 3.2% of source neutrons (3.0 cm non-borated polyethylene and 20.3 cm borated polyethylene), ambient gammas ranged from 0.5% to 8.0% of source neutrons, and source gammas ranged from 2.9% to 23% of source neutrons. The number of counts subtracted was therefore small for cases with less shielding. Even where there was more shielding, a visual inspection of the data showed that the gamma subtraction was quite good as it cleanly removed the gamma region from the ${}^6\text{Li}$ data. As long as all four runs for the same shielding configuration are done consistently, this subtraction along with the number of neutrons identified are not significantly affected by gain nor by the choice of the neutron region boundaries. This is due to the fact that although the peak areas of Fig. 2 scale with details of gain and boundaries, the number of counts in the neutron region remains the same. Some neutrons in the low-peak-area tail of the distribution may not be counted; however, that number is very small compared to the total number of neutrons.

It is also important that we accurately model the detector-source system in the Monte Carlo code. We included the lithium glass, the lucite, the aluminum can in which the detector is housed, and steel sheets below the detector and below the source in the Monte Carlo simulations. To the extent that room return from other sources is important, it must be modeled correctly as well, but it is more difficult to enter accurate details of the lift and the underlying concrete. Using MCNP, we estimated the concrete and the lift to add less than 2% to the number of neutrons detected.

One other possible source of systematic error is the diurnal variation in cosmic-ray neutron events [1]. We took all four runs for each data point in immediate succession so as to minimize diurnal variation. Note that it is not appropriate to take ${}^6\text{Li}$ and

^7Li runs simultaneously due to detector cross-talk, including radiation from one photomultiplier tube producing a signal in the second tube.

Lastly, the source activity has a 3.2% uncertainty at the 99% confidence level.

The most significant sources of systematic error are then: room return, which could add as much as 2% to the measured neutron count rates, and uncertainty in the source activity. Together, they are not likely to account for the discrepancies that we observe between MCNP simulations and experiment.

4. Conclusion

We constructed a pair of ^6Li and ^7Li glass detectors expressly for the purpose of comparing MCNP simulations of shielded neutron sources to experimental data. This detector system could detect fission neutrons from ^{252}Cf with up to 24 cm of non-borated polyethylene or 20.3 cm of borated polyethylene shielding with counting rates well over the neutron background rate. Both non-borated polyethylene and borated polyethylene give an initial increase in neutron detection efficiency as many neutrons are thermalized, but a large number are still able to escape the shielding without being captured. As shielding is added, the detector's practical intrinsic efficiency quickly decreases as more and more neutrons are captured and never escape the shielding. Numerical values are given in Table 1.

[Table 1]

MCNP does quite well at modeling non-borated and borated polyethylene, but has a general tendency of underestimating detector efficiency. For non-borated polyethylene, it underestimates the measured value by an average of 8%. This increases to an average of 11% for borated polyethylene.

It is difficult to separate the contributions of the detector and of the shielding in the discrepancies observed, but it does appear that MCNP overestimates neutron absorption in thick polyethylene while possibly shifting the neutron spectrum to lower energies than observed. Experimental data for additional detectors would help sort out the source of these discrepancies between the simulation and the experiment.

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364 Fig. 1. Detector geometry.

365

366 Fig. 2. Distribution of pulses in the ^6Li (circle) detector and the ^7Li (triangle) detector as a function of pulse area. The data are
367 for 3.0 cm of non-borated polyethylene shielding above and below the source.

368

369 Fig. 3. MCNP calculation of the percentage of lithium capture taking place in each sheet of glass as a function of shielding
370 thickness above and below the source.

371

372 Fig. 4. Energy distribution of neutrons leaving non-borated polyethylene (PE) shielding of the thicknesses both above and
373 below the source as indicated in the legend.

374

375 Fig. 5. MCNP calculation and the measured practical intrinsic efficiency for non-borated polyethylene as a function of
376 shielding thickness above and below the source.

377

378 Fig. 6. A comparison of the practical intrinsic efficiency of the ^6Li detector with non-borated and borated polyethylene
379 shielding. The dashed curve is the ratio of the results for borated to non-borated polyethylene. Shielding thicknesses are both
380 above and below the source.

381

382 Fig. 7. MCNP calculation and the measured practical intrinsic efficiency for borated polyethylene as a function of shielding
383 thickness above and below the source.

384

385 Fig. 8. MCNP calculation of the percentage of neutrons absorbed by a ^{252}Cf source placed between two 2.54 cm-thick sheets
386 of 25.4 cm $\times$ 25.4 cm borated polyethylene as a function of the weight percentage of boron in the polyethylene. Boron is
387 added as borate (BO_3) and the polyethylene density is fixed at the measured value.

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Table 1
Practical intrinsic efficiency of the ^6Li detector for a ^{252}Cf source shielded with non-borated polyethylene and 5% borated polyethylene.

Non-borated Polyethylene Shielding (cm)	Practical Intrinsic Efficiency (%)	Borated Polyethylene Shielding (cm)	Practical Intrinsic Efficiency (%)
0.0	7.6	0.0	7.6
3.0	52.5	2.5	26.6
6.0	40.9	5.1	20.7
9.0	22.8	7.6	13.8
12.0	12.4	10.2	8.4
15.0	6.6	12.7	4.8
18.0	3.5	15.2	3.2
24.0	1.4	20.3	1.3