MONTE CARLO SIMULATIONS OF NEUTRON MULTIPLICITY COUNTING FOR
INTERNATIONAL NUCLEAR SAFEGUARDS

By

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MONTE CARLO SIMULATIONS OF NEUTRON MULTIPlicity COUNTING FOR
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By

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MCNP simulations are used to improve efficiency in the design and understanding of multiplicity and coincidence counters. However, there are many assumptions and uncertainties in the simulations. Reducing these uncertainties and enabling MCNP to closely predict measurements would improve efficiency in design, testing, calibration, and operation, increasing the International Atomic Energy Agency’s confidence in verifying compliance with the Nonproliferation Treaty. While resolving all the uncertainties would take a large collaborative effort, this work addressed four areas of uncertainty.

The ability of MCNP to simulate cosmic rays was investigated by comparing simulations with measurements of cosmic ray’s effect on multiplicity counters. The accuracy of the simulation was quantified and the simulation was used to calculate cosmic ray properties that would be impossible to measure, such as the contribution to the singles, doubles, and triples as a function of cosmic ray particle type. The results of these calculations are being implemented into cosmic ray rejection techniques.

AmLi sources are used in active interrogation neutron coincidence measurements, but the AmLi spectrum is not well known. There is significant variation

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between different AmLi sources due to different manufacturing methods. Seventeen AmLi sources were measured to characterize the variation in spectra and ideal spectra were generated based on these measurements. The spectra allow MCNP users to quantify and reduce their uncertainty.

MCNP has traditionally simulated fission by table lookups of average neutron signature values. New fission models FREYA and CGMF have been developed and incorporated into MCNP6.2, which simulate detailed physics of individual fission processes and generate correlated neutrons. These models were investigated in simulation of multiplicity counters and were found to correct some significant assumptions made in MCNP6.1.1b, namely the neutron angular correlation. However, the models incorrectly simulate some basic neutron properties, such as the Cf-252 energy, and need further work.

Finally, the effects of a few millimeters of metal source encapsulation was characterized as it is often dismissed as neutronically ‘light’. The effects of various materials and thicknesses were quantified and the encapsulation required for a Cf-252 source to match the efficiency of Pu-240 in an Active Well Coincidence Counter was found.
CHAPTER 1
NEUTRON COUNTING FOR INTERNATIONAL SAFEGUARDS

International Safeguards

Historical Motivation

The discovery of nuclear fission in 1938 was accompanied by the realization that large quantities of energy were released through the reaction on the order of 100 million times greater than a chemical reaction. It was quickly realized that fission could cause a chain reaction which could be used in reactors and weapons, and in August 1939, Albert Einstein sent a letter to President Roosevelt suggesting investigation into such a weapon and warning of Germany’s actions towards the same [1]. On July 15th, 1945, the United States’ Manhattan Project successfully detonated the world’s first nuclear weapon in the desert of New Mexico, and 30 days later World War II ended with Japan’s surrender.

Nuclear technology proliferated as a military secret with Russia and then the United Kingdom testing their first nuclear weapons. Then President Eisenhower’s “Atoms for Peace” speech in 1953 directed a shift in the application of nuclear technology towards peaceful civilian purposes [2]. The speech led to the formation of the International Atomic Energy Agency (IAEA), tasked with promoting atomic energy for peaceful purposes while limiting military applications. Nuclear weapons continued to proliferate, with France and China testing nuclear weapons by 1964.

Nuclear weapons radically influenced the balance of power. Much of the international structure that exists today originated because of nuclear weapons. NATO and many international relationships are due to the nuclear umbrellas of the US and Russia. The permanent members of the UN Security Council, and those with veto
power, are the first five countries to acquire nuclear weapons. Several countries have pursued nuclear weapons as a means to defend against the influence of other countries and there are many examples of military action to prevent countries from acquiring nuclear weapons. Preserving their influence is a strong incentive for the current nuclear weapons states to encourage non-proliferation.

It is also in the best interests of non-weapons states to prevent proliferation. The world is safer without nuclear weapons. The more countries have nuclear weapons, the greater chance one would be used either by accident or purposefully and the greater chance more would be used in retaliation. These incentives led to the creation of the Non-Proliferation Treaty (NPT), the international framework to prevent the spread of nuclear weapons. Countries agree not to acquire nuclear weapons and peaceful nuclear technology is safeguarded against diversion to nonpeaceful purposes by the IAEA. The treaty came into effect in 1970 [3].

The NPT provides a mechanism for countries to trust that their neighbor is not pursuing nuclear weapons, and so their own incentive is reduced. Openly demonstrating through IAEA inspections that they are not pursuing nuclear weapons also provides strong standing to pressure other countries into adopting IAEA safeguards. Removing the incentive and providing pressure to not acquire nuclear weapons influences the decision making of countries such that pursuit is no longer in their best interest. This effect is especially potent because the NPT is the most signed treaty in the world with 191 signatories. Research has shown that this framework has significantly reduced the probability that states will pursue nuclear weapons [4]. In addition, several countries have given up nuclear weapons programs, including South
Africa, Brazil, and Argentina. Still, the NPT failed to stop India, Pakistan, and North Korea from acquiring nuclear weapons.

The non-nuclear weapons states received concessions in exchange for relinquishing proliferation. The NPT has two more ‘pillars’ in addition to the nonproliferation of nuclear weapons; the disarmament of nuclear weapons states and the peaceful use of nuclear technology. The disarmament of weapons states was a key reason non-weapons states agreed to the NPT. The NPT language is vague, requiring parties to pursue disarmament in good faith. Non-weapons states argue disarmament has not happened fast enough, and in response created the Treaty on the Prohibition of Nuclear Weapons, passed in July 2017 [5]. The US and Russia are the focus of disarmament, as their arsenals dwarf any other state’s. They counter that their arsenals are on a continuous decline and over 90% of their weapons have been eliminated.

The final pillar, the peaceful use of nuclear technology, has grown the use of nuclear science for energy, industry, research, and medicine around the world. The IAEA continues to support this aspect and also facilitates international cooperation on nuclear safety, with an increased focus after the Fukushima disaster. However, while this nuclear technology is being used peacefully, the spread of nuclear material throughout the world has greatly increased the risk of theft. Research reactor cores of highly enriched uranium could be used in a non-state actor’s nuclear weapon and large medical sources could be used with conventional explosives as a ‘dirty bomb’. Work is ongoing to minimize this risk by removing excess nuclear material in all countries.
Safeguards Regime

Article III.1 of the NPT requires non-weapons states to accept safeguards, ‘as set forth in an agreement to be negotiated and concluded with the IAEA in accordance with its Statute and the Agency’s safeguards system’. The structure and content of that agreement is detailed in Information Circular (INFCIRC) 153 [6] It states that a national system of material control and accounting (MCA) shall be established such that the IAEA can verify ‘there has been no diversion of nuclear material from peaceful uses to nuclear weapons’. Safeguards will be applied towards this purpose ‘on all source and special fissionable material in all peaceful nuclear activities’. Obligations regarding IAEA inspectors are described. In addition to the measures described in INFCIRC 153, ‘subsidiary agreements to specify in detail how procedures are to be applied’ are described. Subsidiary agreements can be implemented at a specific facility level for example. The IAEA Statue further describes that the inspectors ‘shall report any non-compliance to the Director General who shall thereupon transmit the report to the Board of Governors. The Board shall call upon the recipient State to remedy any non-compliance. The Board shall report the non-compliance to all members and to the Security Council and General Assembly of the United Nations. The United Nations is the enforcement body to violations of the treaty while the IAEA’s role is limited to reporting.

INFCIRC 153 describes the scope of inspections to include inspecting records, measuring material, verifying the functionality of instruments, applying surveillance and containment, and other technically feasible methods. Nondestructive assay, and specifically neutron multiplicity counting, is a small part of IAEA safeguards. The
majority is seals, cameras, and verifying that items and facilities are being used as declared.

Note that thus far only the verification of declared activities has been described. This safeguards regime was proven ineffective by the discovery of Iraq's undeclared efforts to develop nuclear weapons in 1991. The revelation of a nuclear weapons program in South Africa, who had not signed the NPT, in 1989 and the detection of North Korea's, who had signed the NPT, noncompliance with safeguards in 1993 brought further attention to the issue. In response the Additional Protocol (AP) was created as INFCIRC 540, with a goal of strengthening safeguards [7]. The AP is a voluntary agreement which 129 states have brought into force. The AP expanded the IAEA’s obligations from verifying declarations to also verifying the absence of undeclared nuclear material and activity within a state. The AP expanded safeguards to include the full fuel cycle, adding mining and storage of waste. It also strengthened the use of environmental sampling and satellite imagery and introduced complimentary access, allowing inspectors to access many identified locations to assure the absence of undeclared nuclear material and activities.

Safeguards Technology

A variety of technology is used to verify the safeguards regime [8]. Cameras provide continuous monitoring for the majority of the time when inspectors can not monitor in person. They allow verification of facility records and monitor equipment and materials for undeclared use. Seals or tamper indicating devices (TIDs) allow continuity of knowledge (CoK). Inspectors can seal their own equipment to ensure it is not compromised until they return and they can seal locks, doors, or equipment to ensure
they have not been accessed. 3D scans of rooms are compared to design information to detect, for example, changes in piping that could indicate undeclared activities in a facility. Environmental swipes of walls, air handling systems, and equipment can detect miniscule amounts of hard to clean particles from the presence of materials. Outside of facilities open source information such as news reports or scientific articles are analyzed. Satellite imagery is also used.

Nondestructive assay by measuring radiation emission is also performed [9]. These techniques take advantage of radiation emitted by materials of interest in nuclear safeguards. The radiation signatures of these materials are unique. Faking them would be incredibly difficult, expensive, and in many cases impossible. Measuring radiation signatures is one of the most robust attribute verification methods available. Handheld gamma spectroscopy systems like the HM-5 are used to verify the presence of specific isotopes, verify the active length of fuel rods, and measure the enrichment of uranium. The Cerenkov light emitted by slowing down of beta particles emitted by spent fuel in water is measured to verify spent fuel ponds. The Irradiated Item Attribute Tester also verifies spent fuel by measuring gamma radiation, while the FORK detector does the same by measuring both gammas and neutrons. Neutrons are also measured through coincidence or multiplicity counting in relatively rare cases when the fissile mass of a sample needs to be verified. This is most common where HEU exists, or where plutonium is processed, such as in Japan where it will be implemented at the Rokkasho reprocessing plant [10].

**Neutron Multiplicity Counting**

For further information on the topics of this section see the Passive Nondestructive Assay (PANDA) manual [11].
The neutron primary interaction mechanism at fission energies is to scatter. Scattering deposits some energy following collision kinematics where energy and momentum are conserved. Neutrons typically scatter many times and travel large distances relative to gamma rays. Neutrons would not deposit their full kinetic energy in a typical detector sized object and are not emitted at discrete energies, so neutron spectroscopy is rarely performed. Instead neutron counting is used, where neutrons scatter until they reach thermal equilibrium and are ‘slow’ and then are absorbed in capture reactions. The resulting energy is on the order of 1 MeV, is mostly deposited in the detector, and clearly indicates one neutron was detected. Neutron energy information is not preserved. Counting the number of neutrons is used to indicate the amount of neutron emitting material present.

More information is needed however, as two sources of identical fission mass can have different neutron emission rates based on their chemical composition due to $(\alpha,n)$ interactions and based on their multiplication, dictated by moderation, geometry, density, and similar considerations. The timing between the neutrons is used as this additional information. Multiple neutrons detected at the same time indicate they came from fission, unlike $(\alpha,n)$ neutrons which are emitted one at a time. Measuring doubles or pairs of neutrons, two neutrons at the same time, is called coincidence counting. Coincidence counting is typically done when the neutron emitter is well known. When more characteristics of the emitter are unknown, the rate of triples or three neutrons at the same time can be measured for additional information. This is called multiplicity counting. Measurements of uranium samples only use coincidence counting while plutonium measurements use coincidence and multiplicity counting depending on how
well the source is characterized. Some of the research presented in this work is relevant only to coincidence counting while some applies to both methods. When both are relevant typically multiplicity is referred to, but if coincidence counting is the more likely application it may be referred to instead.

There exists some probability that two uncorrelated neutrons will be detected at the same time because they accidentally overlap. These neutrons could come from a random source such as AmLi or a correlated source such as two independent spontaneous fissions occurring at the same time. Neutron counting cannot distinguish from where a neutron was emitted so there is no way to tell if two detections at the same time came from a correlated source or not. However, statistically, on average, the rate of detecting accidental coincidences can be found. The detected coincidence rate consists of real and accidental coincidences from which the rate of accidental coincidences is subtracted. This leaves the quantity of interest, real coincidences or ‘doubles’, which correspond to the mass of fissioning material. Accidental and real coincidences are often referred to as ‘accidentals’ and ‘reals’. Subtraction of accidentals is required in coincidence and multiplicity counting.

There are many ways of analyzing the timing between detected neutrons. This discussion begins with Rossi-Alpha, the theoretical analysis that best explains the concepts involved [12]. Then the analysis actually performed by most detector electronics, called shift registers, is described [13]. Finally, Feynman-Y analysis, a similar technique that is rarely used in safeguards, is described [14]. These methods essentially all give the same results and are different ways of organizing the information contained in the timing between neutrons.
The Rossi-Alpha distribution is formed by measuring the time of detection of neutrons after some arbitrary starting neutron. If that starting neutron is the first neutron detected from a fission event then more neutron detections will quickly follow. If the starting neutron is from a random event like \((\alpha, n)\), the next detected neutron is equally probable at any time because it is independent of the starting neutron. Plotting the number of detected neutrons at a given time after the starting neutron will produce Figure 1-1 [11].

![Figure 1-1](image)

Figure 1-1. Rossi-alpha distribution. Photo credit: Reilly [11].

Random uncorrelated neutrons have a constant probability at all times and make a horizontal line on the plot. Correlated neutrons from the same fission event or chain are most likely close in time to the first neutron, creating the sloped feature. The slope follows the shape \(e^{(-t/T)}\), where \(t\) is the time after first detection and \(T\) is the dieaway time. The dieaway time is an important characteristic of a detector. It describes how quickly the neutron population in the detector dies off following a fission. The neutron population falls off as neutrons are detected (captured in He-3 tubes), escape, or are absorbed. The dominant impact on this time is how far thermal neutrons can travel before being absorbed or detected. In this chapter the concept of ‘at the same time’ has been used. A more accurate description is ‘close together in time’. The dieaway time
describes how close together neutrons from the same fission or fission chain will be. Gate widths are then set to include most of the neutrons from a fission chain. The gate width is the length of time chosen to be considered 'close together' and from the same fission chain. Frequently 64 microseconds is used.

The plot also illustrates the subtraction of accidental coincidences. Two gates are used, G on the plot. The first gate, close to t=0, captures real and accidental coincidences. Then a long delay, D, is used to be certain that no neutrons from a fission at t=0 remain. Typically 1024 microseconds is used which is many dieaway times. The second gate captures only accidental coincidences. The gate lengths are equal. Because accidental coincidences are constant with time the rate of accidentals in each gate will be the same even if an individual coincidence cannot be identified as real or accidental. Subtracting the rate of accidental coincidences leaves only the real coincidences which indicate the fissioning material mass.

The final characteristic illustrated by the plot is the predelay, P. The shape of the curve shows an increase from t=0 to a short time after, where the number of events peaks and then starts to fall. This is because when a neutron is detected there is electronics deadtime that makes any He-3 tube connected to it ‘dead’. This effectively causes the detector to have a lower efficiency for a short period of time. Historically 4.5 microseconds has been used, but recent fast electronics can lower this to 1.5 microseconds. If the first gate opened at t=0 it would get a lower accidentals count rate than the second gate because for part of the first gate the detector has a lower efficiency. The predelay ensures both gates have equal accidentals count rate.
Most multiplicity counting systems use shift register electronics to apply these principles. Neutron detections should be visualized as a pulse train, a stream of pulses indicating the detection time of a neutron. As time increments the pulses ‘flow’ through the electronics. The shift register diagram is shown in Figure 1-2 [11]. As a pulse enters the electronics it triggers the R+A scaler to record how many pulses are coincident with the first pulse. Then it enters the shift register and the counter is incremented by one. Any additional neutrons detected during this time will be coincident with the first neutron. After the gate length has elapsed the first pulse exits the shift register and the counter is decremented by 1. The first pulse will no longer be counted as a real coincidence by any other incoming pulses. After a long delay the A scaler adds the value of the counter, recording the number of pulses accidentally coincident with the original pulse. The shift register method allows recording of coincidence of every incoming neutron with every neutron in the gate even if the gate from the previous neutron has not ended. In effect every neutron opens its own gate without the deadtime of waiting for the previous neutron’s gate to finish.

Figure 1-2. Shift register flow diagram for R+A gate. Photo credit: Reilly [11].
The Feynman-Y or Feynman variance method is another analysis technique of neutron correlations. It was first used to perform reactor noise analysis in the 1940s. The count rate is recorded in randomly started gates of the same time width. If the source is random the distribution will follow Poisson statistics and the variance-to-mean ratio will be 1. When correlated neutrons are detected this quantity will be greater than 1, and can be used in the same way that the R count rate is in shift register analysis. Cifarelli and Hage expanded this analysis to three parameters [15].

**Multiplicity Counter Design**

Gas proportional counters filled with He-3 are the most common detection mechanism in neutron counters. Other thermal neutron capture materials such as boron or lithium are used with limited effectiveness. Alternate technologies have lower neutron capture cross sections and can have difficulties detecting the reaction energy, reducing detection efficiency. He-3 is especially preferred for its stability and is known to operate for decades in facilities without requiring maintenance. He-3 systems are stable with temperature and time. They do not deplete as the neutron capture reaction produces tritium which decays back to He-3. It is a mature technology with decades of refinement and institutional knowledge which is attractive in nuclear facilities where safety, stability, and regulation are key. He-3 was generated from tritium decay from the weapons program, which produced an excess leading to availability and widespread adoption. With the decline of the weapons program in recent decades its production is restricted and a full He-3 tube costs about $3,000. The impact is apparent in modern detectors with over 100 tubes.
With respect to high voltage the tube is operated just above the knee in the proportional region, typically about 1700V. As the reaction products move through the tube they ionize the gas inside creating electrons and positive ions. The high voltage field pulls the electrons into the center anode wire, causing additional ionizations proportional to the number initially created by the reaction. The electrons are collected at the center wire making a voltage pulse. This pulse is typically much larger than could be deposited by a gamma ray due to the low Z and density of the gas. Gammas are easily discriminated. First a preamplifier shapes the signal, then it is amplified, and then a discriminator discriminates gamma pulses. A TTL pulse is generated for each neutron capture which is read by the shift register. The described circuit is the source of most system dead time, and improvements to the electronics is an active field of research [16]. Typically multiple He-3 tubes are connected to a single amplifier board. To preserve uniform count rates and dead time across the amplifiers, the number of tubes per amplifier depends on their expected count rate. The inner ring of tubes where count rates are highest might have 3 tubes per detector while the outer ring has 7 tubes.

Thermal counter design involves thermalizing or slowing the neutrons and then capturing them. Fission neutron energy of 2 MeV must be reduced by 8 orders of magnitude to 0.0253 eV where capture cross sections are highest. Moderation is most effective with hydrogen with an equal mass to neutrons. Polyethylene, -(CH₂)-, is used as an inexpensive, workable, stable source of hydrogen. Detectors are typically He-3 tubes imbedded in polyethylene. The tube spacing and polyethylene thickness is chosen to maximize efficiency and minimize dead time. The carbon and, mostly, hydrogen in polyethylene will capture thermal neutrons before they reach He-3 tubes in
a process known as parasitic capture. This reduces the counting efficiency. While some polyethylene will increase count rates as more neutrons are thermalized, too much will lower count rates as the rate of parasitic capture exceeds thermalization. Typically the detectors are under moderated or too little polyethylene is used as the sample itself will cause some moderation and regularly contains low Z materials such as water or oxygen.

This lead to the use of multiple rings of He-3 tubes. The inner ring is then able to absorb neutrons that were quickly thermalized while high energy neutrons more likely to penetrate further into the detector before thermalization are absorbed by outer rings. Of course the additional cost of He-3 tubes is considered. The most advanced multiplicity counters have four rings. The addition of multiple rings also added a new capability to thermal neutron counters, a crude measurement of energy. Higher energy neutrons are more likely to reach the outer rings so taking the ratio of counts between rings indicates neutron energy. This is similar to the application of a Bonner sphere which has long been used to measure neutron energy.

Design considerations affect the dieaway time. A detector figure of merit (FOM) is efficiency squared over dieaway time, Equation 1-1. While originally moderator was used to spread out the neutrons in time so they would not all fall within the predelay before the gate opened, it is typically desired to minimize the dieaway time. The statistical uncertainty on the real coincidences, found from the R+A – A gates, increases as 2A, Equation 1-2. Selecting the gate width is a tradeoff between capturing more reals at the expense of more accidentals. Minimizing the dieaway time allows the gate width to be decreased while the same amount of reals are captured, reducing uncertainty and
improving FOM. A typical gate width is 64 microseconds while state of the art detector
gate widths are as low as 24 microseconds. The dieaway time and neutron lifetime is
dominated by thermal neutrons traveling at 2,200 m/s, which is relatively slow compared
to when they are emitted at 100,000,000 times higher energy. Minimizing the amount of
time neutrons spend as thermal will minimize the dieaway time. Neutron absorbers
perform this function. The biggest dieaway driver is a neutron being emitted from the
sample, thermalizing in the polyethylene, and returning to the sample, inducing fission
which can then repeat the process. Wrapping the detector cavity in a neutron absorber
like cadmium will significantly reduce the dieaway time. Cadmium can also surround the
He-3 tubes, ensuring that any thermalization happens inside the tubes where absorption
and detection is immediate, although this comes at the expense of reduced efficiency.
This technique is paired with high fill pressures of He-3, making detection of neutrons at
low but not yet thermal energies more likely. Other designs involve moving the tubes
close together to reduce the distance thermal neutrons can travel before being
absorbed. Cadmium liners on the outside of the detector reduce dieaway time while
also reducing background counts.

\[ FOM = \frac{\varepsilon^2}{\tau} \]  \hspace{1cm} (1-1)
\[ \sigma(R) = \sqrt{(R + A) + A} \]  \hspace{1cm} (1-2)

Uniform detection efficiency within a source and within a sample cavity is also a
priority. Multiplicity counters for safeguards are most often well counters, where a
sample cavity or ‘well’ sits in the center of the He-3 tubes. If the source is placed closer
to the left tubes it will be further from the right tubes and the count rate will be
unaffected. The same applies to fissions on the left side of the sample. Graphite
reflectors which plug the sample cavity from above and below perform the same function on the vertical axis by reflecting neutrons in those directions towards the He-3 tubes. This design allows the measurement system to robustly operate with tolerance for typical fluctuations found in industrial facilities.

There are often high radiation backgrounds in nuclear facilities that must be accounted for. This is reduced by increasing the polyethylene thickness on the outside of the detector. This acts as a shield that reduces the background count rate. Cadmium liners are used in conjunction to absorb neutrons once they have been moderated or before they enter the detector. This design consideration must be balanced with size. The best performing detectors are large enough to moderate and capture most neutrons, and additional polyethylene for shielding causes their weight to be several hundred pounds. Caster wheels allow movement by a team of people. However these detectors are normally left at a facility between uses. Where portability is required smaller detectors are used, weighing tens of pounds and with attached briefcase handles.

**Application to Plutonium and Uranium Measurements**

Neutron multiplicity counters are often designed for a specific application to optimize and account for limitations in what is being measured. Some common applications are discussed.

Uranium is measured as UF$_6$ in enrichment facilities, as an oxide powder in bulk processing, and as fuel assemblies. U-235 is the isotope of interest for its applicability to nonpeaceful purposes. Uranium has miniscule spontaneous fission rates making it difficult to measure passively. Instead active interrogation is used. AmLi is the most
common active interrogation source with a maximum neutron energy of about 500 keV, well below the 1 MeV ‘threshold’ for fission in U-238. This ensures that all measured coincidences come from U-235 fission. The U-238 spontaneous fission rate is negligible in comparison and is ignored. Random sources are used as interrogators because they do not contribute to the coincidence rate. Uranium is almost never analyzed with multiplicity counting, only coincidence counting. The use of interrogation sources greatly increases the accidentals count rate degrading the statistical uncertainty of the triples rate beyond usability. Of the three unknowns, mass, (α,n) rate, and multiplication, often multiplication is negligible or can be accounted for in calibration curves so only the doubles must be measured.

Recall that the cadmium liner of a sample cavity ensures that no thermal neutrons enter the cavity. At low masses of uranium the cadmium liner will be removed, greatly increasing the thermal flux, the number of fissions, and the coincidence count rate. At larger uranium masses thermal neutrons can no longer penetrate the sample and the count rate no longer changes with mass. For large uranium masses the cadmium liner must be used ensuring only fast neutrons are in the cavity. These fast neutrons can fully penetrate the uranium. While the count rate is reduced because only fast neutrons or neutrons thermalizing inside the uranium induce fission, the interrogation is uniform and the result is robust.

The Active Well Coincidence Counter (AWCC) is designed to measure cans of uranium [17]. A picture and MCNP model of the detector are shown in Figs. 1-3 and 1-4 [18]. In the first figure a shift register is shown on the table. An insert to measure MTR fuel assemblies is next to the detector. In the MCNP model a can of uranium sits in the
cavity. Lime green is air, blue is polyethylene, teal is uranium, and the AmLi source is in orange. Although not in this model, typically a second AmLi source is placed in the top plug for uniform interrogation. A typical measurement time is 10 minutes. The Uranium Neutron Coincidence Collar (UNCL) is designed to measure fuel assemblies [19, 20]. It comes in PWR and BWR configurations. It is shown in Figure 1-5 and an MCNP model is shown in Figure 1-6. Orange is air, blue is He-3 tubes, and yellow is AmLi. A typical measurement time is 30 minutes, and it can identify the diversion of about 5 fuel pins. The door holding the AmLi source slides out of the UNCL so the remaining 3 sides can be slid around the assembly. Note that the detector is asymmetrical with only 1 AmLi source. Fuel pins closest to the interrogation source are furthest from the He-3 detectors creating a uniform interrogation profile.

Figure 1-3. AWCC, shift register, and MTR fuel measurement insert. Photo credit: Kouzes [18].
Figure 1-4. MCNP model of the AWCC. Photo credit: Weinmann-Smith.

Figure 1-5. UNCL with fuel assembly. Photo credit: Kouzes [18].
So far only count rates have been discussed, not calculations of fissile mass. The most common analysis method is the simple calibration curve. Samples of known mass are measured and their doubles, coincidence, or reals rate are plotted as a function of mass. An equation is fit to the curve. Then the coincidence rate of unknown samples is recorded and the equation is used to find their mass. An example coincidence curve is shown for the AWCC in Figure 1-7 [17]. Calibration curves perform best when the unknown samples closely match the calibration samples. This applies to geometry, chemical form, container, and any other characteristic that would affect the count rate. This is typically ensured through administrative controls and unexpected results are investigated. Mass is also a limitation. An unknown larger than the largest calibration sample should not be measured. The low alpha decay rates and multiplication of uranium assist in robustness of the method. Some physics effects affect the shape of the curve. For uranium, self-shielding causes the slope of the curve to decrease at high masses. However multiplication causes the slope to increase at higher masses. For plutonium the multiplication effect is much stronger and the curve increases
exponentially. Calibration curves are always applied to uranium and can be applied to plutonium that is well characterized.

Figure 1-7. AWCC calibration curve. Photo credit: Menlove [17].

The other analysis methods applied to plutonium are known alpha and multiplicity. Unlike uranium, plutonium sources are strong alpha emitters and have high multiplication and these effects must be treated rigorously. If the plutonium chemical form is known and controlled then alpha, the ratio of \((\alpha, n)\) rate to spontaneous fission rate, can be known. Then the effects of multiplication from spontaneous and \((\alpha, n)\) neutrons can be subtracted and a calibration curve can be used. Singles and doubles are measured to solve for the two unknowns, mass and multiplication. A calibration curve without this correction, even if the \((\alpha, n)\) rate is constant, would be incorrect as samples with the same mass will have different multiplications and different doubles rates.
To perform the known alpha correction first a nonmultiplying sample with known alpha should be measured and the singles and doubles rates $D_0$ and $S_0$ recorded. Alpha can be calculated from the sample isotopics and chemical form. The isotopics are verified through gamma measurement. Then the unknown multiplying sample should be measured and the singles and doubles rates recorded. Alpha for the unknown sample must also be calculated. A parameter $r$ is calculated by Equation 1-3, which is coupled with the multiplication $M$ to form the correction factor. $M$ is calculated from Equation 1-4. The measured doubles rate $D_m$ is then corrected in Equation 1-5 to give $D_c$. $D_c$ depends only on the spontaneous fission rate and a calibration curve of $D_c$ is compared to $D_m$ in Figure 1-8.

$$r = \frac{D/S}{D_0/S_0} \frac{(1+\alpha)}{(1+\alpha_0)} \quad (1-3)$$

$$0 = 2.062(1 + \alpha)M^2 - [2.026(1 + \alpha) - 1]M - r \quad (1-4)$$

$$D_c = \frac{D_m}{Mr} \quad (1-5)$$

Passive and known alpha calibrations are the most common methods of assay. Multiplicity counting is used only rarely, and only for plutonium. It requires high efficiency detectors because the probability of detecting a triple scales with efficiency cubed. It uses measured singles, doubles, and triples with an analytic model to solve the three unknowns, mass, multiplication, and alpha. It requires the least information known about the sample, requiring no calibration standards and being robust with respect to impurities and changes in multiplication. However because the triples scale with efficiency cubed it has the poorest statistical uncertainty, requires longer measurements, and performs best with high activity samples. Multiplicity counting uses
the point model equations which show the singles, doubles, and triples rates expected based on detector parameters, nuclear data, and the three sample unknowns. When the singles, doubles, and triples are measured, software uses the equations to solve for the three unknowns. The point model equations follow. The singles equation is the fission neutron emission rate scaled by the multiplication, alpha, and detection efficiency, Equation 1-6. The doubles rate, Equation 1-7, is the rate of detecting two neutrons at the same time. It includes the fission emission rate, spontaneous fission neutrons inducing fission, and (α,n) neutrons inducing fission. It also includes terms for the fraction of doubles collected in the gate, which would be 1 for an infinite gate. It is divided by 2! to account for double counting, where both neutrons A and B and neutrons B and A count as doubles. The triples rate, Equation 1-8, includes terms for the different combinations of neutron production in the sample [21].

Figure 1-8. Example calibration curve before and after known alpha correction. Photo credit: Weinmann-Smith.
\[ S = F\varepsilon MV_s(1 + \alpha) \] (1-6)

\[ D = \frac{F\varepsilon^2 f_d M^2}{2} [v_{s2} + \left(\frac{M-1}{\nu_{i1}-1}\right) v_{s1}(1 + \alpha) v_{i2}] \] (1-7)

\[ T = \frac{F\varepsilon^3 f_t M^3}{6} [v_{s3} + \left(\frac{M-1}{\nu_{i1}-1}\right) [3v_{s2} v_{i2} + v_{s1}(1 + \alpha) v_{i3}] + 3 \left(\frac{M-1}{\nu_{i1}-1}\right)^2 v_{s1}(1 + \alpha) v_{i2}^2] \] (1-8)

Where

\( f = \) spontaneous fission rate
\( \varepsilon = \) neutron detection efficiency
\( M = \) neutron leakage multiplication
\( \alpha = \) \((\alpha, n)\) to spontaneous fission neutron emission rate ratio
\( f_d = \) doubles gate fraction
\( f_t = \) triples gate fraction
\( v_{ij} = \) average number of \( j \) neutrons produced in \( i = \) spontaneous or induced fission

Plutonium has 5 isotopes of concern, complicating measurements compared to uranium assay. Pu-238 is uncommon because of its production mechanism. Pu-239 is most common and is generated from neutron capture and double beta decay of U-238. Pu-240 thorough Pu-242 are produced from subsequent neutron capture in Pu-239. Their population increases with burnup, and Pu-240 will be the most common. The even numbered isotopes emit about 1000 times more spontaneous fission neutrons per second per gram than the odd numbered isotopes. Still, the Pu-239 spontaneous fission rate is greater than U-238. Neutron assay cannot distinguish which isotope a detected neutron was emitted from. Because of this plutonium assay finds the Pu-240 effective mass, or the amount of Pu-240 that would generate the measured spontaneous fission
rate. It is effectively assumed that all of the neutrons come from Pu-240. The increased spontaneous fission rate per gram of Pu-238 and Pu-242 is taken into account by Equation 1-9. Gamma measurements are used to find the sample isotopics. These isotopics are used once the Pu-240 effective has been found to calculate the mass of all the isotopes in the sample.

\[ Pu_{eff}^{240} = 2.52Pu^{238} + Pu^{240} + 1.68Pu^{242} \]  
(1-9)

A diagram of the high level neutron coincidence counter (HNCC-II) is shown in Figure 1-9 [22]. It is a single ring detector used to measure plutonium with calibration curves. Note the increased polyethylene thickness on the top and bottom to smooth the efficiency profile. The mini epithermal neutron multiplicity counter (mENMC) is a state of the art neutron multiplicity counter for multiplicity analysis of plutonium [23]. It is shown with an MCNP model of the detector in Figure 1-10 and Figure 1-11. It can split open for analysis of irregular samples.

Figure 1-9. Diagram of the HLNCC. Photo credit: Menlove [22].
Limitations and Active Research

This section is a brief summary of limitations and active research in the field of neutron counting for safeguards, including topics addressed in this work.

Cosmic rays are a particularly difficult source of background neutron coincidences [24]. High energy cosmic rays interact in the atmosphere generating
showers of high energy particles. These particles interact in walls, detectors, samples, and floors causing neutron spallations with the same time correlation as fission. The sample itself, often dense and high Z, generates many of these fissions, so traditional background subtraction by measuring without the sample is ineffective. The random nature of cosmic rays further complicates analysis. Improved cosmic ray subtraction would reduce the minimum detectable activity of low mass samples, leading to great cost savings and improved capabilities in decontamination and decommissioning.

The reduced production of He-3 has prompted investment in alternative technologies [25]. While there is no longer a projected shortage of He-3, these technologies could reduce cost and prevent a shortage in the future. The technologies almost all center on some thermal neutron absorber like boron or lithium and some detection mechanism. BF3 tubes are proportional counters similar to He-3. Lithium glass acts as a scintillator and is used with photomultiplier tubes. Semiconductor detectors have been investigated. While many individual advantages have been identified, no candidate has clearly demonstrated better total performance over He-3. Replacements have lower cross sections and poor charge collection.

Scintillators are a unique potential replacement being aggressively pursued by the IAEA [26, 27, 28]. They have been investigated in prior decades as well [29]. Instead of capturing thermal neutrons they measure scintillation light from fast neutron recoil on hydrogen. These systems have dieaway times on the order of 0.1 microseconds, allowing incredibly short gate widths compared to thermal systems. The resulting chance of an accidental count, overlap between two independent events, is much smaller when the time window is over 100 times smaller. This greatly improves
statistical uncertainty and reduces measurement times, especially in applications where the accidentals rate is high such as active interrogation of uranium. They can measure gamma radiation as well which increases the available signatures for analysis. They have not moved beyond the testing phase because of poor stability and complicated analysis. They are temperature dependent which can bias analysis. They are most often made of a toxic flammable liquid which prohibits deployment in nuclear facilities. It is unclear how they perform after decades of use in the field and after sitting for up to a year at a nuclear facility between uses. They are sensitive to gamma pulses and rely on pulse shape discrimination to identify neutron detections. This method requires large neutron energy thresholds and intense analysis which limits the detector performance. Their efficiency is low. If these problems can be resolved the benefits of fast neutron detection can be taken advantage of.

List mode analysis appears to be the next iteration on shift register technology [30]. Instead of analyzing the pulse train as it arrives and then discarding it, list mode refers to recording the detection time and location of every neutron pulse in a list. The data can then be reanalyzed with different methods, gate widths, and other parameters. The ability to store neutron detection location, sometimes down to the individual He-3 tube, makes available new information and analysis techniques to improve multiplicity counting.

There is still room for improvement to basic characteristics of detectors. While the most obvious improvements, more He-3 tubes, are limited by detector cost, some others are still available. Improvements to preamplifier electronics are continuous, reducing deadtime and the necessary predelay. The great performance improvements
of fast multiplicity counters could be approached by minimizing the lifetime of thermal
neutrons in a detector. Reducing the predelay further is a necessary first step to making
this viable. As the dieaway time shrinks, the count rate lost to predelay grows.
Placement of neutron absorbers at points furthest from detectors would reduce the
dieaway. The use of graphite instead of polyethylene eliminates parasitic neutron
capture at the expense of greater volume needed for the same amount of moderation.
Although probably too expensive, the use of heavy polyethylene would also improve this
factor. Implementation of these improvements would be slow. The incremental
performance increase would not be worth the cost of new procedures and production
methods and so these improvements are not pursued.

The uncertainty of source characteristics is another significant limitation. Monte
Carlo tools like MCNP are used to simulate detectors. A completely new detector can
be ‘built’ and tested with any source in an afternoon for free. Of course, MCNP is not
perfect. It is limited by the accuracy of its nuclear data. In general it can reach answers
within 5%-10% of measurements. One of the biggest limitations to improving its
performance is the accuracy of a source’s neutron emission rate. A NIST calibration of a
Cf-252 source reports the answer with a 1-sigma uncertainty of 1%. MCNP reports its
results per starting source particle and it must be told the source strength to compare
with measurement results. An MCNP result with 0 statistical uncertainty is still +2% at
2-sigma from the source strength alone, making it impossible to know whether most
changes to MCNP have improved the results or not. Accurately knowing the ‘right’
answer MCNP should reach is crucial for reducing uncertainty and improving MCNP
performance.
MCNP and the Monte Carlo Method

The Monte Carlo method is a tool to simulate complex systems. Monte Carlo N-Particle is a nuclear physics simulation program which uses the Monte Carlo method [31]. The framework of MCNP began in the Manhattan project and has been maintained and developed ever since, making it one of the oldest programs in existence. It is a repository of modern nuclear physics knowledge. It combines particle transport and probabilities of events to simulate almost any geometry. The method involves picking random numbers which dictate what happens based on their probability functions. It is primarily used to model critical systems for reactor analysis and criticality safety applications. This discussion focuses on applications in neutron multiplicity counting.

An MCNP example is described. First a 3D model of a detector is built, specifying what space each material occupies. A Cf-252 source at the center is specified. A spontaneous fission neutron is generated, its energy picked randomly with the probability of the Cf-252 neutron energy spectrum. Its direction is also picked randomly, and it ‘travels’ through the detector while a random number decides how far it travels before interacting. This distance is based on the cross section of the material it travels through. When it interacts a random number decides what interaction occurs. There is a 20% chance of capture, a 60% chance of scatter, and a 20% chance of fission, and the random number is between 0 and 1. These chances represent cross sections, nuclear data that have been carefully measured. If the random number is less than 0.2 then the neutron was captured, less than 0.8 and it scattered, and less than 1 and it induced fission. What happens next is chosen randomly based on the probabilities of those events and so on until the neutron is terminated. This process happens for millions or billions of simulated neutrons and the probabilities of some event happening, say, how
often fission is induced or how often a He-3 capture occurs, is recorded. With enough samples every possible combination of events in a highly complex system can occur with enough frequency to make a statistical determination of their probability. In a detector the singles rate or probability of He-3 capture per starting neutron is found. Almost any quantity can be tallied. A carefully modeled simulation can give information that would be impossible to measure experimentally. With modern computers a detector is fully simulated in hours or minutes.

Physics options can affect what data and methods are used in different parts of the simulation. Fully simulating every detail would greatly increase the calculation time so the options are also used to eliminate aspects of the problem that are unimportant. For example, photons can generate electrons through the photoelectric effect. The code can simulate this interaction as the photon depositing energy in the material and terminating the photon or it can create electrons with this energy which are themselves transported through the simulation. For gamma detectors this is rarely performed. Similarly, a simulation of a fast neutron detector may use a neutron energy threshold where particles that fall below a certain energy are terminated because they no longer have the kinetic energy to register above the real detector’s detection threshold. Physics options are also chosen based on what data should be used. Specific cross section data may be preferred for performance or historical reasons. A certain high energy neutron interaction model may give better answers for one application over another.

These options lead to a serious trap in the results of the simulations. It is quite easy for two simulations of the same scenario to use different options and give different results. It is easy to choose the wrong options to get a wrong answer and it is easy to
assume the code is simulating some aspect of the simulation when it really is not, or is making some simplification that gives incorrect results for the application. The MCNP manual, already 763 pages long, is notoriously limited in its explanations. In the earlier example of a neutron termination threshold eliminating neutrons below the threshold for a fast neutron detector, the results could be incorrect because lower energy neutrons still undergo capture reactions that generate more neutrons or generate photons which fast neutron detectors can detect.

Even when MCNP is used perfectly the results do not match experiments exactly. The random natures of both radiation measurements and Monte Carlo simulations mean results from both have uncertainties. Further uncertainties in source neutron emission rates, geometry specifications, and especially nuclear data cause measurement and simulation to diverge. Agreement must be defined based on the application. An MCNP result 4% away from a measurement with a 5% acceptance criteria is acceptable, while being 4% away from reproducing a simple Cf-252 measurement with a 1-sigma uncertainty of 1% indicates the simulation is inaccurate. It also sheds doubt on application towards the 5% measurement. While the simulation agreed this time, something is clearly wrong and perhaps next time it will not agree. Agreement is also defined based on the uncertainties of the nuclear data and physics models. Reviewing documentation of data in a typical multiplicity simulation will reveal 10% uncertainties on (α,n) yields, spontaneous fission yields, neutron energy spectra, and transport and interaction cross sections. Smaller but significant uncertainties are listed on the remaining data. It is then curious that simulations nearly always agree much better than these uncertainties would suggest. This could be attributed to a lucky
counterbalancing of effects, but is most likely due to overestimations of uncertainty and updated data. Also there is no centralized and uniform method of uncertainty quantification and uncertainties are often unreported.

Reducing these uncertainties will converge on ‘perfect’ Monte Carlo. The results will never match exactly but 0.1% accuracies are an example. Such improvements would enable capabilities which increase efficiency and cost savings in executing the safeguards mission. Currently MCNP is used extensively in safeguards detector research. Almost every study uses it to test design changes, predict results, and calculate difficult-to-measure information. However it is always benchmarked to measurements and almost never used for absolute rates. MCNP is used to show that adding another He-3 tube will increase the count rate by 3%, not 100 counts per second. The actual source must be measured with the actual detector and the measured count rate scaled up 3%. MCNP accurately reproducing measurements would save tremendous effort and resources. A country switching to uranium carbide fuel or 20x20 BWR assemblies requires millions of dollars and years of effort in the fabrication of standards to generate the calibration curves needed for assay. Accurate MCNP could finish the analysis in a month. Testing a new detector for spent fuel would no longer require a nuclear facility willing to deal with visitors and their equipment.

While some improvements require wide collaborations, measurements of many materials over years and evaluation of the produced data, there is some low hanging fruit available. Currently so much is uncertain that it is difficult to identify a single target for improvement. A simulation is inaccurate because 10 things are wrong, not just
because of one thing. A path forward is unclear, starting everywhere at once is impractical.

An extremely well characterized measurement is needed. Characterizing an experiment could be readily performed with one or a few people. The resulting highly physically accurate simulation would completely remove uncertainty introduced by the geometry and materials of the measurement. While this sounds easy it is rarely performed because other uncertainties make the act unnecessary for direct applications. The densities of the materials of the measurement should be measured exactly. Polyethylene has a range of density over 10% which changes the detector efficiency as much. In current simulations the polyethylene density is often guessed at.

Once the physical aspects are simulated exactly any remaining differences between simulation and measurement are due to physics simulations and nuclear data. A simple experiment incorporating the best known physics interactions and best known nuclear data should be chosen. For example, Cf-252 is well studied and should be used as the radiation source. This simple detector would narrow the scope of necessary improvements to Cf-252, polyethylene, and He-3 data and neutron transport physics. Once the simulation closely matches the measurements all of the nuclear data is validated. Minor changes to the measurement, one at a time, can be made to validate additional data until it is clear which things are known precisely and which are not, and also to assist in improving knowledge of the unknowns. This would result in accuracy in MCNP simulations that could be used for independent absolute simulations of results.
CHAPTER 2
OBJECTIVES OF THIS WORK

The objective of this work was to improve the capability of neutron multiplicity counting to verify the Nonproliferation Treaty. This was done by addressing shortcomings in data, processes, tools, and knowledge of the technique. Most of the improvements focused on MCNP simulations, but there was some application directly to measurements. This objective was accomplished and the state of the art of multiplicity counting has been advanced in application towards low activity measurements, active interrogation with AmLi sources, fission models in MCNP simulations, and calibration with surrogate sources.

The first area of study is cosmic ray background. As described in previous sections, cosmic rays are a complex source of correlated background neutrons that are difficult to subtract. When high total mass but low activity samples must be measured cosmic rays are the limiting factor in the measurement uncertainty. Developing a robust cosmic ray subtraction method would greatly improve the performance of coincidence counting toward this application. MCNP simulations of cosmic rays are an especially important tool for this problem because of the difficulty of detailed cosmic ray measurements due to the variety of particle types and energies. In this work the accuracy of MCNP simulations of cosmic rays in multiplicity counting was found by comparing simulations to measurements. Close agreement between simulation and measurement was demonstrated and quantified. Because MCNP accurately modeled cosmic rays it was used to calculate data that would be difficult to measure, like the flux and contribution to singles, doubles, and triples by particle type. Data from these results
were used by another student to develop a cosmic ray rejection algorithm, although the performance was lacking.

In the next focus area, assumptions regarding active interrogation of uranium were addressed. AmLi (α,n) sources are used to assay U-235 mass in bulk samples and fuel assemblies. AmLi sources are created by mixing Am and Li based powders. The quality of the mixture and composition of the powders dictates the alpha particle energy lost in transport and the resulting neutron spectrum. This causes the AmLi spectrum to differ by manufacturer and from source to source. While this fact was known, for lack of a better option researchers often picked their favorite AmLi spectrum from the literature and assumed their source had the same spectrum. Proper calibration accounts for the spectrum in measurements but MCNP simulations are unable to use this technique and risk using an incorrect spectrum. In this work 17 AmLi sources were measured in a 5 ring multiplicity counter. The ring ratios were used to indicate the source energy. The variation between source spectra was characterized for different manufacturers and for individual sources. This allows researchers to characterize the uncertainty introduced by their choice of spectrum for the first time. Then a regression fitting method was used to find the spectrum that most closely reproduced the measurement results in MCNP for individual sources and for the average for a manufacturer. The publication of these spectra provides researchers with a spectrum that will more closely match their source because it was generated by an average of sources manufactured like theirs and provides the data to quantify the expected uncertainty by using this spectrum. Finally, all known spectra in the literature were simulated and compared to these measurements to identify the best performers. This
In the third study fission models available in MCNP were compared to quantify their effect on simulations of multiplicity counters in order to guide users towards the best model. In the latest version of MCNP, to be released in 2018, new fission models CGMF and FREYA fully simulate individual fissions by modeling the fission fragments conserving energy and momentum. Neutrons and photons are generated in these simulations with greater correlations between particles from a single fission. This is in contrast to the standard model traditionally used in safeguards simulations which samples a single neutrons’ properties completely independent from other neutrons of the same fission. In this work the properties of the neutrons, their energy, multiplicity, and direction were compared to measured values. Then the effects on a coincidence counter due to changes in these properties were quantified. This was done by simulating simplified detectors which isolated only a single property. For example, a 4π thick detector is only sensitive to changes in multiplicity. The effects on simulations of common safeguards detectors was found by comparison to measurements in the AEFC and AWCC. The results showed that no model clearly outperforms the others. The work quantifies the limitations of MCNP as a safeguards tool and guides the focus of future improvements.

The final study involved encapsulating a neutron source to change the emitted neutron behavior to match another source. The first could then act as a surrogate for the second. Some calibrations of safeguards detectors are often performed at facilities that do not have the SNM which the detectors are designed to measure. More common
Cf-252 sources are used as a surrogate, but the differences in neutron energy between that and Pu-240 for example must be accounted for. Instead of a computational adjustment the Cf-252 source could be encapsulated in some material causing it to have the same detector response as a Pu-240 source. This would prevent mistakes and save time for these calibrations. In this work encapsulations of copper, steel, and polyethylene were manufactured in 4 thicknesses. A Cf-252 source was measured and the change in detector response was found. These results were used to validate MCNP simulations of the measurements which were then used to understand how the neutron spectrum changed with the encapsulation. MCNP was used to calculate the neutron spectrum and detector response to common encapsulations, demonstrating the effects of encapsulation used by different source manufacturers. MCNP simulations of the detector response to different sources and the encapsulation required for a Cf-252 source to have an identical detector response to a Pu-240 source for a common safeguards detector, the AWCC, were also performed. The work demonstrated the encapsulation required to use Cf-252 as a surrogate source. It also quantified the effect of different encapsulations, guiding users in decisions on the level of detail required in simulations to accurately model measurement effects. These results improve understanding for simulations of multiplicity counters.

**Related Accomplishments**

During the completion of these projects additional work was performed. Teaching was a significant and enjoyable portion of effort. Teaching experience included instructing neutron and gamma measurements at LANL’s NDA Fundamentals course. Design and instruction of curriculum for international and university safeguards courses was also performed. The results of the cosmic ray project were used to guide a project
for a summer student at LANL. These and other events have been fantastic exposure to a wide range of NDA practitioners from government, industry, and academia.

Another project involved the characterization of a novel fast scintillator detector. Scintillators detect fast neutrons which enable great advantages in counting times and statistical uncertainties. Scintillators typically have low Z and low density so photons rarely deposit their full energy. This scintillator had a unique ability to fully detect photons, generating photopeaks in a gamma spectrum, because it was doped with tin, a high Z material. This work involved finding the optimal operation characteristics such as operating voltage. The performance in common detector metrics was quantified, including efficiency and resolution. These fundamental results provide a basis for design of large inexpensive detectors for homeland security applications.

An application focused project was an MCNP design optimization study for the Unattended Cylinder Verification Station, UCVS. The UCVS is undergoing years of prototype design and testing for deployment to a safeguards program. It measures the U-235 content and total uranium mass of UF₆ cylinders used for transportation, storage, and processing of uranium during the enrichment process. It uses a gamma detector and neutron coincidence counter which each independently assay the uranium mass and U-235 content. The gamma detector measures enrichment by gamma peaks and measures uranium mass through neutron capture. The detector is wrapped in layers of steel and polyethylene. Neutrons emitted by the sample thermalize in the polyethylene and are captured in the Fe-56, depositing between 3-8 MeV of energy in the detector. This signal is taken as the neutron count rate and thus mass. The neutron counter is two coincidence counters in slab geometry. Singles indicate the U-235 mass and
doubles the U-238 mass. UF₆ cylinders are especially difficult to assay because the material has a triple point at temperatures readily reached during their outside storage. Depending on the weather the material can either be in solid, liquid, or gas, and is likely in some combination of all 3. This form causes the material to solidify on the walls in heterogeneous distributions within the cylinder. This causes the measured signal to be dependent on this distribution and detector placement. This work used MCNP simulations to model the detector response to different distributions of the uranium contents. It also simulated various detector placement options, characterizing performance. Sensitivity to nearby cylinders as background sources was quantified. The detection efficiency profile in 3D throughout the cylinder was visualized. The results guided the design of the UCVS and quantified expected uncertainties due to the fill profile of the cylinders. This work improved the performance of the UCVS, which will eventually reduce counting times and facilitate verification of the Nonproliferation Treaty when the detector is implemented.
CHAPTER 3
COSMIC RAY EFFECTS

Introduction

In low activity measurements background radiation significantly increases statistical uncertainty, biases results, and dictates minimum detectable activity. Background sources of neutrons are rare. Background coincidence neutrons are even rarer, and so measurements of neutron coincidence and multiplicity are relatively insensitive to background neutrons. However, cosmic rays are a background source of coincidence neutrons that is especially difficult to characterize, control, or subtract. They are especially impactful during low activity measurements of uranium. The signal is so small that active interrogation of U-235 is rarely used as results would have poor statistics. The U-238 spontaneous fission neutron yield is 0.014 n s$^{-1}$ g$^{-1}$ [21]. In typical applications such as decommissioning and decontamination the sample mass is a majority of steel or some dense material which produces large amounts of cosmic rays. This in combination with low uranium masses means the sample is often at or below the minimum detectable activity. Better understanding and accounting of cosmic rays will reduce their impact and lower the minimum detectable activity of detectors.

An example of a high energy cosmic ray producing a large number of neutrons through spallation with a dense material is shown in Figure 3-1. Multiple coincident neutrons are created in this non-radioactive material and can then be detected, greatly increasing the minimum detectable activity for neutron emitting materials.

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In this work MCNP6 was used to calculate the cosmic ray contribution to a multiplicity counter measuring 66% enriched uranium, lead bricks, and an empty detector. The results were compared to measurements to determine their accuracy. Comparisons to literature values of some results were also made. These comparisons quantify the accuracy of the simulations which were then used to calculate information difficult to measure experimentally.

Before particle accelerators cosmic rays were the primary source of high energy particles for physics study. Many Manhattan Project scientists studied cosmic rays, including Bruno Rossi of the Rossi-alpha distribution [32]. They determined that the source of cosmic rays are from galactic events, pulsars, stellar flares, galactic nuclei explosions, and supernovas. They are 87% protons, 12% helium nuclei, and 1% heavier nuclei. They have an average half-life of 200 million years. At the edge of the Earth’s atmosphere they are isotropic due to various magnetic fields. Only particles with energy greater than about 1 GeV will reach the surface because as they move through the atmosphere they lose energy through interactions with the Earth’s magnetic field and
through collisions. These collisions produce high energy particles, mostly pions, muons, nucleons, electrons, and photons, which can also have very high energies and create more generations of particles in further collisions. These collisions create cosmic ray showers, large numbers of particles reaching the surface close together at the same time [33, 34, 35].

While the sun influences galactic cosmic rays, its own particles, about 98% protons, lack the energy to penetrate the earth’s atmosphere. The sun’s activity influences the strength of the magnetic field around Earth. The magnetic field increases with solar activity and cosmic rays require more energy to penetrate the magnetic field. The solar cycle affects the surface cosmic ray intensity by up to 30%. The cycle lasts 11 years and was at a peak during this work in May 2014 [36].

Location also affects cosmic ray intensity. Altitude affects the mass of attenuator, air, cosmic rays pass through before reaching the surface. Altitude is reported as g cm$^{-2}$ to account changes in pressure. During this study Los Alamos, NM averaged about 810 g cm$^{-2}$. The magnetic field is strongest at the equator, so at high latitudes cosmic rays penetrate further and the intensity is higher. All of these factors cause great variation in the measurement of cosmic rays making them difficult to predict. The location and solar strength must be accounted for in cosmic ray measurements. Even after accounting for these things cosmic rays vary randomly.

**Measurements and Simulations**

The mini Epithermal Neutron Multiplicity Counter was used for the experiment [23]. The detector uses traditional shift register electronics where counts in the accidentals gate are subtracted from the reals plus accidentals gate. Cosmic ray induced spallation neutrons contribute real coincidences. The detector’s low dieaway
time and high efficiency make it well suited for multiplicity measurements where the factorial moments of the measured multiplicity distribution, singles, doubles, and triples, are measured and used to calculate the three sample unknowns, alpha, multiplication, and mass. While the detector can split open, it was operated closed for this study, Figure 3-2. It has 104 He-3 tubes at 10 atmosphere pressure. The tubes are arranged in four rings. The efficiency is 61.8% and the die-away time is 19.1 microseconds.

![Mini Epithermal Neutron Multiplicity Counter](image)

Figure 3-2. Mini Epithermal Neutron Multiplicity Counter [23].

Two lead bricks were measured with a total mass of 22.6 kg. The lead acts to amplify the cosmic ray intensity. It is a dense high Z material, similar to uranium and plutonium, without spontaneous fission and \((\alpha,n)\) reactions. The uranium sample was UISO-66 [37]. It is 1 kg of uranium in the form of U3O8 with a U-235 mass of 655 grams. It is encapsulated in a thin tinned-steel container.

The MCNP6 simulation used a date of April 2013, 15 months before the measurements were taken. The effect of this difference is negligible for the purposes of this demonstration as the solar activity was very similar during these times, Figure 3-3. Still, for real applications the simulation should be performed at the correct date.
The simulation was performed in several steps to minimize repeated calculations and increase flexibility. First the transport of cosmic rays through the atmosphere was performed. It was created for another LANL project [38]. The source term was the galactic cosmic ray spectrum incident on the earth’s atmosphere at an altitude of 65km. It is given by the integral 2pi flux from Castagnoli and Lal as corrected by Masarik & Reedy with Clem’s rigidity cutoffs [38, 39, 40]. Physic models were used except where ENDF table data exist for neutrons, protons, and photons. The Vavilov model was used for charged-particle straggling. CEM030.3 and LAQGSM03.03 transported charged particles, heavy nuclei, and neutrons and protons [41]. The default models were used in all cases except for replacing ISABEL with LAQGSM03.03 for light ions below 940 MeV/nucleon to reduce calculation time [42].

The source location was specified to be Los Alamos, latitude 36 N and longitude 110 W, and the date was April 1\textsuperscript{st}, 2013. The simulation was 2km by 2km in X and Y.
The particles were transported down 62,810 m to the altitude of Los Alamos, 2190 m. A 15 cm thick concrete floor capped the bottom of the simulation to reflect particles back up, simulating the ground. The current of each of the 14 tracked particle types was tallied 2 m above the concrete. The particle energy spectra were recorded in logarithmic bins between 0.01 eV and 1000 GeV and the directions were recorded in 9 cosine bins, 5 down and 4 up.

This information was the source term for the second stage of the simulation. The particles were emitted at the top of a cylinder with a radius of 50 m. A box the size of the miniENMC was placed at the bottom center. The particles entering the box from the top and averaged over the 4 sides were tallied with the same energy and angle resolution. The cylinder was filled with air and particles that entered the box were killed as to not contribute to the other side.

This was then the cosmic ray source term for a simulation of the miniENMC. This way changes could be made to the detector, for example adding the lead bricks, without simulating the entire cosmic ray atmospheric transport again. The source weights were renormalized to account for the portion of a cosine bin’s particles emitted away from the detector such that the correct number of particles entered the detector. The coincidence capture between the He-3 tubes was tallied with the F8 capture tally and FT8 coincidence special tally treatment to specify the 1.5 microsecond predelay and 64 microsecond gate length. The MCNP6 model of the miniENMC is shown in Figure 3-4.
Results

The measured multiplicity distributions for the empty, lead, and uranium samples are compared to the simulations in Figs. 3-5, 3-6, and 3-7. The overall agreement is good. The results suggest MCNP6 overestimates higher multiplicities. The multiplicity distribution is a probably of each multiplicity being detected. Summing the difference between the two methods for each multiplicity suggests the general agreement between them, accounting for the reduced frequency of larger multiplicities. The total difference of the lead sample was 5.2%, of uranium was 2.3%, and of the empty detector was 1.3%. The relative statistical error of each multiplicity is plotted and is too small to be seen. It was less than 3% relative for the 4th multiplicities. The MCNP statistical uncertainty is precise, but the measurement method did not allow for an accurate calculation of the uncertainties of the probability of each multiplicity and so Poisson statistics are assumed.
Figure 3-5. Multiplicity distributions of the empty detector. Photo credit: Weinmann-Smith.

Figure 3-6. Multiplicity distributions of the lead sample. Photo credit: Weinmann-Smith.
Figure 3-7. Multiplicity distribution of the uranium sample. Photo credit: Weinmann-Smith.

Table 3-1 shows the singles doubles and triples rates with their relative differences. Higher multiplicities had the greatest difference between measurement and simulation, and they have larger influence on doubles and triples. This agrees with the increased difference between simulation and measurement for doubles and then triples. MCNP consistently over predicts every result, and over predicts the singles rates by about 7 counts per second consistently. The addition of lead demonstrates the effect of cosmic rays. The doubles and triples count rates are greatly increased, even more than when adding uranium. The lead rates are predicted within a factor of 2, which is better than expected for the absolute modeling of a complex and random process. The statistical error is shown in the table, and is negligible for the singles and doubles rates.
Table 3-1. Singles, doubles, and triples rates for each sample configuration. Statistical uncertainty is one standard deviation.

<table>
<thead>
<tr>
<th></th>
<th>Empty</th>
<th>Lead</th>
<th>Uranium</th>
</tr>
</thead>
<tbody>
<tr>
<td>Singles</td>
<td>MCNP</td>
<td>27.63(4)</td>
<td>33.45(7)</td>
</tr>
<tr>
<td></td>
<td>Experimental</td>
<td>20.57(2)</td>
<td>24.07(3)</td>
</tr>
<tr>
<td></td>
<td>Difference</td>
<td>34(0)%</td>
<td>39(0)%</td>
</tr>
<tr>
<td>Doubles</td>
<td>MCNP</td>
<td>1.405(3)</td>
<td>23.1(1)</td>
</tr>
<tr>
<td></td>
<td>Experimental</td>
<td>0.804(9)</td>
<td>12.2(6)</td>
</tr>
<tr>
<td></td>
<td>Difference</td>
<td>75(1)%</td>
<td>89(4)%</td>
</tr>
<tr>
<td>Triples</td>
<td>MCNP</td>
<td>0.637(6)</td>
<td>145 (4)</td>
</tr>
<tr>
<td></td>
<td>Experimental</td>
<td>0.30 (3)</td>
<td>104(18)</td>
</tr>
<tr>
<td></td>
<td>Difference</td>
<td>110(9)%</td>
<td>39(7)%</td>
</tr>
</tbody>
</table>

Comparison to other work in the literature demonstrates the level of simulation accuracy that can be achieved. A singles rate agreement of 10-20% was reached by a cosmic ray model for counting of cargo containers, although details of their calculation method were limited so comparison is difficult [43]. Similar work on a cargo scanner comparing MCNP and measurements found a 7% discrepancy for a Cf-252 source [44].

Literature comparison can also be made to related calculations of this work. These quantities were calculated automatically during the MCNP6 simulation although they are not the main focus. The production of tritium in the atmosphere is one such value. This work simulated 0.323 tritons cm⁻² s⁻¹ while another calculation reached 0.320 cm⁻² s⁻¹ [33]. Their value is an average over location and solar cycle and includes the entire atmosphere to sea level. This work’s result is to only 2190 m and during a solar peak, and both will reduce the value which suggests MCNP over predicts the result. In Masarik and Beer the neutron flux and total at a latitude of 30-40 N, altitude of 810 g cm⁻², and solar modulation which are all similar to this work was reported. Their value of 0.405 neutrons cm⁻² closely matches the MCNP6 simulation of 0.383 neutrons cm⁻². The spectra also show a general agreement, Figure 3-8.
Figure 3-8. Neutron spectrum at 810 g cm$^{-2}$ from Masarik and Beer and this work. Photo credit: Weinmann-Smith.

A comparison of the neutron flux was also made between this work and Hess, shown in Figure 3-9 [45]. The MCNP6 flux is depressed for an altitude of 810 g cm$^{-2}$ which is to be expected for a period of high solar activity which can lower the intensity by up to 30%. At low neutron energies ($<1$ eV) the flux depends on the surrounding environment and the poor agreement is to be expected. In other work FLUKA, a Monte Carlo model, calculated cosmic ray spectra for comparison to measurements [35]. Disagreement was as large as a factor of 10, which is about the same as this work’s difference to Hess.

**MCNP6-Derived Cosmic Ray Particle Behavior**

Because MCNP6 was shown to agree with experimental measurements and literature results, the simulation was used to extract information that would be difficult to measure experimentally. MCNP6 calculated neutron detections by the multiplicity counter by particle type and source.
Figure 3-9. The MCNP6 neutron flux at 810 g cm\(^{-2}\) superimposed on the experimental neutron flux Hess values. Photo credit: Roesler [35, 45].

The incidence on the mini-ENMC by particle type is shown in Figure 3-10.

Neutrons and photons are the most intense components of the source, and neutrons will create a direct signal in the detector. They can also cause spallation, generating further neutrons. As a validation of the result, at sea level neutrons should be 30 times more intense as protons and pions, and muons should be 1000 times intense than pions [34]. The results match these estimations. The most common particles have less than 1% standard deviation. The least common particles are sampled the least and have the worst uncertainty because fewer detections are tallied.
Figure 3-10. MCNP6 estimation of particle incidence on the outer surface of a miniENMC at Los Alamos on April 1st, 2013. Photo credit: Weinmann-Smith.

The lead sample’s singles, doubles, and triples were shown by incident particle type. For example, incident protons are responsible for a triples rate of 45. Figure 3-11 shows the calculations. The largest contribution to coincidence counts come from neutrons and protons. A large triples rate relative to the singles rate shows that many neutrons are produced at the same time per total neutrons produced. This indicates large spallations. Positive pions are one such example, and their large spallations are due to an energetic exothermic reaction with matter.

For the uranium simulation the detected multiplicity distribution was calculated for cosmic rays, spontaneous fission, and the (α,n) source individually. These simulations only generated source particles of one type, whereas the full simulation generated each particle individually and then combines the results. The neutron yields for spontaneous
fission and $(\alpha,n)$ were found with SOURCES4C [46]. The $(\alpha,n)$ source only generates a multiplicity of 1, but greater than 1 can be detected because of induced fissions in the uranium. Cosmic rays and spontaneous fission can also induce additional fissions, multiplying the neutron population in the detector. The detected multiplicity by source is shown in Figure 3-12. Note the y-axis range of 7 orders of magnitude. The detected multiplicity from spontaneous fission and $(\alpha,n)$ decrease quickly, large multiplicities are unlikely. This is especially true for $(\alpha,n)$ neutron contributions. Cosmic rays are the majority of the high multiplicity contribution to the detector, which may be useful in developing cosmic ray rejection techniques.

![Graph of particle type vs. rates (s^{-1})](image)

**Figure 3-11.** Absolute singles, doubles, and triples rates by particle type in the lead sample. Photo credit: Weinmann-Smith.
Discussion

The difference between simulations and measurements come from inaccuracies in MCNP6. This work and the related measurements began 15 months after the date simulated in MCNP6. Due to the required time the simulation cannot be redone. However, we have shown the solar activity was at the same level at both times. Although the difference is expected to be negligible, the specified longitude of the MCNP6 simulation was 160 km west of Los Alamos. Inaccuracies in the physical description of the detector, samples, and floor will also contribute. The detector model includes polyethylene, He-3 tubes, and the plugs and sample cavity, but smaller details like bolts, electronics, wheels, and structural supports were neglected. There is also several percent uncertainty in the polyethylene density. Standard concrete was used in the MCNP6 calculation, the actual floor material was unknown and could be different. Particle transport and cosmic ray interactions may differ in these materials, especially

Figure 3-12. Detected multiplicity of the uranium simulation by source. Photo credit: Weinmann-Smith.

Rates (s⁻¹)

Multiplicity

Cosmic rays
Spontaneous fission
(α,n)
when they are high-Z and high density. However, we estimate that the largest source of error in the simulation is physics models. At the high energies and exotic particles of cosmic rays experimental cross sections rarely exist. Physics models are used instead, and often come with large uncertainties relative to experimental cross sections.

Another difference in the MCNP simulation is the correlated particles of cosmic ray showers. MCNP simulates the history of each starting event independently from the others. While cosmic ray showers may generate multiple time-correlated particles, this effect was not simulated in MCNP due to the structure of the runs. Breaking the simulation into multiple runs to maximize efficiency removes time correlation between any particles. Due to the low count rates the effects of this treatment should be negligible, which is supported by the fact that the calculations matched the experimental results reasonably well. Similarly, accidental coincidences are not simulated in MCNP. Only the reals are simulated. Accidentals must be subtracted from measurements before they can be compared with simulation.

The MCNP6 derived cosmic ray behavior illuminates some aspects of cosmic ray detection. One method of cosmic ray rejection is to use muon paddles above the detector in coincidence veto. Charged muons depositing energy in the detector is taken as an indicator of a cosmic ray shower. The detector signal is rejected for some period of time while the cosmic rays pass through. Figure 3-10 shows that muons are the 3rd most common particle and Figure 3-11 shows they induce the 3rd most neutron detections. While they indicate showers, if a single cosmic ray or shower without muons occurs a significant portion of cosmic rays may still be detected.
In Figure 3-10, the high neutron incidence and the counts from the empty detector suggest that a significant background contribution is due to cosmic ray neutrons being detected before they interact with the sample. Since the cosmic rays vary with time taking a background for subtraction would be ineffective and more active cosmic ray suppression techniques may be needed.

Finally, Figure 3-12 shows that, for the uranium sample, when 7 or more neutrons are detected it is most often from cosmic rays. The detected multiplicity of spontaneous fission events decreases quickly as a function of multiplicity. It is possible also that the ratio of low multiplicity to high multiplicity cosmic rays can be calculated. As an example, there could be 100 cosmic rays events with a multiplicity of 1, and 60 with a multiplicity of 2, and so on for every multiplicity of 7 detected. An algorithm could be developed to take the measured multiplicities of 7 or more, assuming they are all cosmic rays, and subtract the expected multiplicities of 1-6, which are a combination of cosmic rays and source events that cannot be discriminated from each other. It is possible the likelihood of multiplicities greater than 6 is so low that the subtraction will have too poor statistics to be practical. The ratio of each multiplicity may have to be adjusted for the source material, uranium may generate more 1-3 multiplicity cosmic rays than plutonium for example. But because the technique is applied through the cosmic rays that are actually measured, it should be robust to changes in location, time, and the random nature of cosmic rays.

**Conclusions**

Neutron coincidence and multiplicity signals of low activity samples can be washed out by background neutrons caused by high energy cosmic rays. This limits the lower detectable sample mass which reduces the applicability and increases cost.
Traditional background subtraction is ineffective, so MCNP6’s cosmic ray feature was investigated as a tool for developing other cosmic ray rejection methods. The code was demonstrated to accurately simulate cosmic rays especially considering the complexity and the use of modeled cross-sections in the system. An average difference in singles rates of 31.3% was found when comparing measurement and simulation of lead, uranium, and an empty detector. The lead singles, doubles, and triples absolute rates were predicted within a factor of two and other particle fluxes and productions agreed with literature values. The results show that the MCNP6 cosmic ray feature can accurately model cosmic rays.

Because MCNP6 closely matched experimental values, it was used to find information that could not be measured experimentally. This information will be useful in cosmic ray rejection techniques. The particles incident on the detector and their contributions to the singles, doubles, and triples rates were found, showing that many of the cosmic ray contributions are single neutrons. This information is key for rejection methods involving shielding or detector design. The coincidence count contribution by source from \((\alpha,n)\), spontaneous fission, and cosmic rays was shown for each multiplicity in a sample of uranium. This revealed that high multiplicities are almost exclusively from cosmic rays while at low multiplicities cosmic rays are still the majority. This information can be used to develop a rejection algorithm based on the probabilities of high multiplicities occurring and their relationship to low multiplicity cosmic ray events.

Several options will be explored in the future. Different high-energy physics options in MCNP6 will be evaluated to investigate the error from the physics models used. Measurements and MCNP6 calculations will be performed in the future at
different points in the solar cycle to evaluate MCNP6 at different solar activities. Other locations will also be utilized to account for different altitudes, latitudes, and longitudes. Ultimately, the MCNP6 capability and information shown here can be used in future studies to develop cosmic ray rejection techniques that reduce the lower detectable mass and reduce uncertainty in low activity coincidence counting.

**Application in Other Work**

The results of Figure 3-12 were applied to a cosmic ray rejection algorithm by Mr. Taylor Harvey. His results are summarized here to show the conclusion of some of the proposed future work for the cosmic ray project [47].

The objective was to develop an algorithm to subtract the detected neutrons from cosmic rays. This is done by measuring the number of times 6+ neutrons are detected in the coincidence gate. It is assumed that this high multiplicity is only due to cosmic rays for low multiplying samples. Then, the cosmic ray contribution to 0-5 neutrons in the gate is calculated from the ratio to the 6+ contributions. For example, if 100 counts in the 1 bin are expected for every 6+ bin, and the 6+ bin detects 0.5 counts per second, then 50 counts per second are subtracted. The ratio of 6+ to other bins was adjusted for mass and material, and was found through measurements and simulation.

Measurements were taken of a range of lead masses, the UISO-66 source, and combinations of the two. A background measurement was subtracted from the all of the measurements to account for room background and cosmic ray generated neutrons in the detector. The lead measurements had masses ranging from 5.9 kg to 32.9 kg and were used to find the ratios of each bin as a function of mass. MCNP6 simulations of the detector and lead were used to calibrate the correction for different materials. In the
simulation the lead was replaced with many different materials to understand the effects of atomic mass on the cosmic rays.

The effectiveness of the rejection algorithm was quantified by comparing its prediction of cosmic ray contribution to measurements of the UISO-66 source. The expected (α,n) and spontaneous fission emission rates were subtracted from the measurement so that only the cosmic ray contribution was remaining. The singles, doubles, and triples from cosmic rays differed by 82%, 55%, and 61% respectively, with the algorithm underpredicting. This approach is susceptible to large errors. Adjusting the algorithm from lead to uranium is uncertain and based only on simulations, not measurements. The neutron yield from (α,n) and spontaneous fission is known to about 10% uncertainty. Further, the mass of uranium was 1kg which is much lower than the range of lead calibrations, the minimum of which was 5.9 kg.

The effectiveness of the algorithm was tested for another application, accounting for the addition of heavy nonradioactive materials in a sample. The UISO-66 source was measured with and without a lead brick in the sample cavity. This should have little effect on the measured neutrons if cosmic rays can be subtracted. This application is also more robust because the subtraction of lead’s effects are guided by measurement. The results reflect this fact, comparing the cosmic ray contributions predicted by the algorithm and from measurement show an agreement of 9%, 23%, and 32% in the singles, doubles, and triples respectively. In this application the algorithm overpredicts the cosmic ray contribution. This result is closer to the expected agreement. The paper concludes that addressing some of the limiting uncertainties in the project promises to improve the accuracy of the algorithm for its application in the field.
CHAPTER 4
VARIATIONS IN AMLI SOURCE SPECTRA

Introduction

The safeguards verification of uranium is often performed by active interrogation because of its low neutron yield. About 75% of IAEA inspection measurements are to verify declarations of uranium items. Both the Uranium Neutron Coincidence Collar and Active-Well Coincidence Counter are used in these measurements and use active interrogation [19, 17]. In these detectors an americium-lithium (AmLi) source emits neutrons which induce fission in the uranium at a rate much greater than the sample’s spontaneous fission. The coincidence neutrons from these induced fissions are measured and calibration curves allow the calculation of U-235 mass [8]. Such calibrations are built from well-characterized standards. Over time access to historical standards has been reduced or calibrations are required when the manufacture of such standards is too expensive to be possible. Instead, modeling and simulation of the calibrations is increasingly used to overcome this limitation. However, the simulations often employ simplifications where processes are poorly known and relying on these simulations introduces potential biases in the calibrations that were not present before.

One such simplification in simulating active interrogation detectors is the AmLi neutron energy spectrum. Accurately simulating this spectrum is critical to the source interrogation rate and resulting measured observables. The uncertainty in the source spectrum prior to this work represented a large portion of the total simulation uncertainty. Several AmLi spectra have been measured [48, 49, 50, 51, 52, 53, 54, 55,

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A few more have been calculated [57, 58, 59]. To illustrate the shape of the spectra, several are plotted in Figure 4-1. In addition, a spectrum was generated in SOURCES4C, which is a widely used program for simulating $(\alpha,n)$ interactions. That spectrum is shown in blue. The spectra typically have two features from the two methods of generating neutrons. The low energy portion from 0 to 1.5 MeV is due to $(\alpha,n)$ interactions on lithium while the portion at higher energies is due to reactions on oxygen. The GVZ spectrum clearly identifies the difference as it only considers interactions on lithium. However the high energy oxygen tail is important because the U-238 fission cross section greatly increases at about 1 MeV. The intensity of high energy neutrons from oxygen controls the induced fission of U-238. Typically U-238 fission is assumed to be negligible but this may not be accurate based on the oxygen content in the sample.

Figure 4-1. Literature spectra normalized per bin width. Photo credit: Weinmann-Smith.
These studies all measured a single source. A researcher using one of these spectra must assume it is the same as the one generated by the source they will be using. The variations in the spectra clearly demonstrate the likelihood of this assumption which introduces large biases into the simulations. As this work shall show, spectrum differences between sources is important at the accuracy of typical MCNP simulations. However, measurements to quantify this difference and identify a manufacturer-specific or individual AmLi spectrum had not been performed prior to this work.

The generation of neutrons in an AmLi source is a complex and poorly characterized process. The neutron spectrum is dependent on physical characteristics which depends on the manufacturing process. Most sources are decades old, the manufacturing companies are out of business, and the processes were not published. The sources were made by mixing americium and lithium based powders with an atom ratio of about 1:400. The americium was an oxide and the result is a matrix of AmO$_2$ particles surrounded by lithium particles. The alpha particles are generated in the decay of americium and they lose energy as they travel out of the AmO$_2$ particles while also having a chance to undergo $(α,n)$ on oxygen, although the cross section is small compared to lithium. The alpha particles are emitted at about 5.5 MeV and can only lose about 1 MeV before they’re below the interaction threshold on lithium while the threshold on oxygen is much lower. Thus variations in AmO$_2$ particle size change the emitted alpha particle energy which controls what fraction of the emitted neutrons were generated on oxygen vs lithium which in turn dictates the energy spectrum. The excess alpha particle energy also influences the resulting neutron energy. As the particle radius increases beyond the infinite thickness of 10 micrometers the emergent alpha spectrum
remains constant but O(α,n) reactions still increase. Variations in manufacturer processes that change the AmO₂ particle size will therefore impact the neutron spectrum. Mixing, often done with a ball mill, has a similar effect as AmO₂ particles next to each other will still produce neutrons on oxygen.

Another consideration is the chemical composition of the lithium. There is little documentation of this characteristic, but the literature refers to LiH, Li₂O, and LiOH. The chemical form and density affect relative neutron generation by lithium and oxygen. For example, a lithium oxide will generate more neutrons from oxygen because of the increased oxygen presence. The ratio of lithium to americium also has an impact and is poorly known. Manufacturers cite lithium mass but it is unclear if this refers to the mass of lithium only or of the lithium matrix, and it is unlikely that pure lithium is used [60]. An example of the uncertainty is highlighted by the MRC-95 source. The documentation states active dimensions of 14.2mm length by 20.6mm diameter with an ‘isotope weight’ of 0.185 g and a ‘target weight’ of 2.3 g. Additional contamination must also be considered. The glove boxes used in the fabrication of these sources are used for other radioactive sources also. Some studies found up to 0.5% of the neutron emission came from interactions with beryllium, with a much higher average energy than lithium or oxygen [49]. Gamma spectra taken in this work did not indicate beryllium contamination, but a detailed gamma NDA study of AmLi sources would be beneficial. These considerations will vary between manufacturers and also from source to source, and so will the neutron spectra.

The source matrix and the capsule will also affect the neutron spectrum as it is transported through the source. A lithium composition including hydrogen will be an
especially strong moderator for example. AmLi sources are often encapsulated in tungsten shielding to reduce the dose, primarily from Am-241 low energy gamma rays. This also affects the neutron energy. Measured spectra from the literature measure the neutron energy after this transport while calculated literature spectra give the neutron energy as they are born. Comparing the two must account for this effect.

The relevant data to this discussion is shown in Table 4-1 including alpha particle decay energies and (α,n) thresholds for pure thick target isotopes. The average neutron energy is for the natural isotopic composition of an element undergoing (α,n) reactions with Am-241 alpha particles as calculated in SOURCES4C.

Table 4-1. Physical properties of the relevant isotopes [61, 11].

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Natural abundance (%)</th>
<th>Alpha energy [branching ratio] (Mev)</th>
<th>(α,n) threshold (MeV)</th>
<th>Average neutron energy by element (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Am-241</td>
<td>5.486 [0.86]</td>
<td>5.443 [0.13]</td>
<td>5.389 [0.01]</td>
<td></td>
</tr>
<tr>
<td>Li-6</td>
<td>7.5</td>
<td>6.32</td>
<td></td>
<td>0.59</td>
</tr>
<tr>
<td>Li-7</td>
<td>92.5</td>
<td>4.38</td>
<td></td>
<td></td>
</tr>
<tr>
<td>O-16</td>
<td>99.76</td>
<td>15.2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>O-17</td>
<td>0.04</td>
<td>0.00</td>
<td></td>
<td>2.45</td>
</tr>
<tr>
<td>O-18</td>
<td>0.2</td>
<td>0.85</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Be-9</td>
<td>100.</td>
<td>0.00</td>
<td></td>
<td>5.10</td>
</tr>
</tbody>
</table>

**Measurements and Simulations**

The 5 Ring Multiplicity Counter was used as an indicator of the neutron spectrum [62]. The neutron count rate in the 5 rings of the detector are recorded individually, and are dependent on energy. The He-3 tubes are embedded in a polyethylene matrix so penetration into the detector correlates with neutron energy. The ring ratios, each ring count rate divided by the total count rate, are taken to neutralize the uncertainty in the source emission rate.
To quantify agreement of spectra and measurement or of a group of measurements with each other a weighted residual sum of squares (RSS) was used. The RSS was weighted by the relative contribution of each ring such that each ring was weighted equally. It was also weighted by the statistical uncertainty such that more accurate values were given more consideration. The weights were all normalized to the inner ring ratio. In practice the weighting nearly cancels out as the outer ring with low count rate has a weight increase to contribute the same as the inner ring with a higher count rate while the outer ring’s worse statistics reduce its’ weight. The metric is summed over all 5 rings. The RSS is shown in Equation 4-1. In application of the RSS to, say, the Cf-252 sources with each other, the ‘expected’ ratio is the average of all Cf-252 sources while the ‘test’ ratio is for an individual source. In quantifying agreement of spectra with measurements, the spectra are simulated in MCNP6 and the resulting simulated ratio is ‘test’ while the measured ratio is ‘expected’.

\[
RSS = \sum_{r=1}^{5} \left( \frac{test - expected}{relative_{t} \times error_{t} \times error_{e}} \right)^{2}
\]  

(4-1)

To characterize the detector six californium sources were measured. Three were A7-series and three were Cf-series. Californium was chosen because of its well-known and consistent energy spectrum. The RSS between californium sources was used to define ‘good’ agreement and benchmark the agreement of the AmLi sources and of the AmLi spectra. The measurements were also used to benchmark the accuracy of the MCNP6 simulations of the 5RMC.

Then 17 AmLi sources from 3 different manufacturer series were measured in the most comprehensive study of AmLi sources to date. Three Gammatron C, eight
Gammatron N, and six MRC sources were measured. The measurements were taken of individual sources and were also averaged over each series.

The simulations were much more complex. First, the MCNP6-based 5RMC model was generated to match the physical detector as closely as possible. As has been discussed elsewhere in this dissertation, the uncertainty on the polyethylene density is about 2% and a limiting factor in MCNP6 accuracy. The density in the model was adjusted within the uncertainty until the simulation most closely matched the measured Cf-252 ring ratios.

Once the model was physically accurate and reproduced Cf-252 measurements it could be applied to the AmLi measurements. The AmLi matrix chemical form was considered carefully. Li$_2$O, LiH, and LiOH of varying densities were considered and LiOH with a density of 1.6 g cc$^{-1}$ corresponding to 1 particle of AmO$_2$ per 263 particles of LiOH was found to best fit the data and literature. This material was chosen as the AmLi matrix for the simulations. The RSS sensitivity to the other matrix forms was found. Also, the model used MRC encapsulation when simulating MRC source measurements and Gammatron encapsulation when simulating Gammatron source measurements.

Once the 5RMC model and AmLi source were as accurate as possible response functions were generated by recording the detector efficiency by ring as a function of the starting neutron energy. Using the response function allows the evaluation of different spectra without the timely process of running the simulation again.

Two simulations were used to generate the bounds on the possible neutron spectrum. The spectrum was then adjusted within the bounds to match the measurements. First, MCNP6 was used to simulate Am-241 alpha particles transported
through AmO$_2$ with radii from 1 to 7 micrometers. The energy spectrum of the emergent alphas was tallied and used as the input to a SOURCES4C simulation of ($\alpha$,n) interactions on lithium and oxygen. The minimum and maximum intensities of each energy bin from the SOURCES4C simulation were the upper and lower bounds of possible spectrum adjustments to match the measurements. The bounds were extended up and down by a factor of 4 to account for unknown variations and increase the fitting precision.

Next the simulations and measurements were combined to generate the neutron spectrum. The neutron spectrum multiplied by the response function for each ring gives the ring ratios. This is performed for each energy bin and then summed over all bins. The target ring ratios come from the measurements. The neutron spectrum was adjusted within the bounds to minimize the difference between the simulated and measured ratios. The lithium and oxygen spectra were adjusted within their bounds, and the relative intensity of oxygen and lithium was also adjusted. The adjustments were performed iteratively until the spectrum generated ring ratios closely agreed with the measured ring ratios.

The result is spectra generated for average C-series, N-series, and MRC-series, and individual N-437 AmLi sources. The spectra are based on the transport and energy loss of alpha radiation through AmO$_2$ particles which undergo ($\alpha$,n) interactions with some ratio between lithium and oxygen, where the resulting neutrons pass through an AmLi matrix. While individually the matrix composition, AmO$_2$ particle size, relative lithium contribution, lithium and oxygen spectra shape, or 5RMC MCNP6 model may be
incorrect, the spectrum that best fits the measured data on average includes the effects of these unknowns.

Finally, all known literature spectra were also simulated and compared with measurements to identify the best spectra and compare agreements between spectra. The best spectra from this work, measured literature, and calculated literature were compared in a simulation of active interrogation of 2kg of HEU metal to demonstrate the range of results delivered in the assay of SNM mass by even the best spectra, highlighting the importance of this work. The worst literature spectrum was also compared. The doubles rate was converted to mass through a typical calibration curve and the result only shows the sensitivity of U-235 mass to the chosen AmLi spectrum. It does not indicate spectrum performance because a different AmLi source used in the measurements would give different results.

Results

The Cf-252 measured inner ring ratios are shown in Figure 4-2 and outer ring ratios are shown in Figure 4-3. The average across the measurements is shown as the black line and the statistical uncertainty of each measurement is also plotted. Note that the scale is quite small here indicating the similarity between the sources. At this statistical uncertainty their spectra are indistinguishable and the variation between sources is negligible. The outer ring ratios have the lowest count rates and largest uncertainty. The statistical uncertainty and standard deviation of the sources with their average for the inner ring ratio is shown in Table 4-2. The RSS of Equation 4-1 is also shown where the agreement of the greatest outlier source (A7-866 for Cf-252) with the average for all the sources is compared. This RSS is the target agreement for the 5RMC MCNP6 model and the generated AmLi spectra such that the model and the
spectra would agree with measurements better than a set of Cf-252 sources agreed with each other. The inner and outer ring ratios for the AmLi measurements are shown in Figs. 4-4 and 4-5 and their statistics are also included in the table. The ring ratio values are tabulated in Table 4-3. The AmLi standard deviation is 5 times larger than that of californium, demonstrating the increased variation between sources for the inner ring. The statistical uncertainty is also much smaller. The RSS between AmLi sources is given for each of the 3 series and each is much greater than the RSS of the Cf-252 sources.

![Graph showing inner ring ratios of californium sources and their average and statistical uncertainty](image)

**Figure 4-2.** Inner ring ratios of the californium sources and their average and statistical uncertainty. Photo credit: Weinmann-Smith.
Table 4.2. Statistical uncertainty and standard deviation of the inner ring ratios. Agreement of the Cf-252 and AmLi sources.

<table>
<thead>
<tr>
<th></th>
<th>Cf-252</th>
<th>AmLi</th>
</tr>
</thead>
<tbody>
<tr>
<td>Statistical uncertainty</td>
<td>0.11%</td>
<td>0.01%</td>
</tr>
<tr>
<td>Standard deviation</td>
<td>0.12%</td>
<td>0.55%</td>
</tr>
<tr>
<td>RSS (C, N, MRC)</td>
<td>1.4*10^{-6}</td>
<td>1.7<em>10^{-5}, 6.2</em>10^{-5}, 4.1*10^{-6}</td>
</tr>
</tbody>
</table>

Figure 4-3. Outer ring ratios of the californium sources and their average and statistical uncertainty. Photo credit: Weinmann-Smith.
Figure 4-4. Inner ring ratios for the AmLi sources. Photo credit: Weinmann-Smith.

Figure 4-5. Outer ring ratios for the AmLi sources. Photo credit: Weinmann-Smith.
Table 4-3. C, N, and MRC series measured ring ratios.

<table>
<thead>
<tr>
<th>Ratio</th>
<th>C-series</th>
<th>N-series</th>
<th>MRC-series</th>
</tr>
</thead>
<tbody>
<tr>
<td>R1</td>
<td>0.49541</td>
<td>0.49016</td>
<td>0.48964</td>
</tr>
<tr>
<td>R2</td>
<td>0.35387</td>
<td>0.35472</td>
<td>0.35548</td>
</tr>
<tr>
<td>R3</td>
<td>0.11465</td>
<td>0.11704</td>
<td>0.11733</td>
</tr>
<tr>
<td>R4</td>
<td>0.02891</td>
<td>0.03027</td>
<td>0.03007</td>
</tr>
<tr>
<td>R5</td>
<td>0.00714</td>
<td>0.00779</td>
<td>0.00747</td>
</tr>
</tbody>
</table>

The simulated 5RMC polyethylene density that best fit the Cf-252 measurements was 0.953 g cc\(^{-1}\). The simulation did not agree with the measurements to within the RSS of the measured Cf-252 sources, although it did agree within the RSS of the measured AmLi sources. This indicates that the MCNP6 model has some unknown bias which should be considered when using these spectra in simulations of other detectors. The RSS values and the model’s sensitivity to polyethylene density are shown in Table 4-4.

Table 4-4. Accuracy of the MCNP6 simulation of the 5RMC and its sensitivity to polyethylene density

<table>
<thead>
<tr>
<th>Simulation</th>
<th>RSS</th>
</tr>
</thead>
<tbody>
<tr>
<td>5RMC best MCNP model (poly density = 0.953 g cc(^{-1}))</td>
<td>9.0*10(^{-6})</td>
</tr>
<tr>
<td>5RMC MCNP model (poly density = 0.950 g cc(^{-1}))</td>
<td>1.7*10(^{-5})</td>
</tr>
</tbody>
</table>

To simulate the 5RMC for AmLi, the AmLi matrix and encapsulation must be added. The matrix material that best agreed with the data was LiOH with a density of 1.6 g cc\(^{-1}\). This and other material’s plausibility was discussed in other work [63]. The RSS for the C-series spectra generated in this work is shown in Table 4-5. The sensitivity is also shown by simulating the same spectra with different matrices. As a check on the calculation of the ring ratios using the response function, the spectrum
was instead input directly into MCNP and the negligible bias introduced through the
response function was quantified. Then the encapsulation was changed to the MRC
Model 2724-BT encapsulation. Finally the last entry is the effect of removing the
tungsten pig, which all standard sources have to reduce gamma emissions. This
demonstrates the sensitivity of the RSS metric to the 423 g of tungsten.

Figure 4-6 shows the simulated 5RMC response function used in the calculation
of the ring ratios for the AmLi spectra. The simulation matched the measurement
conditions which were the Gammatron model AN-HP-9 encapsulation in a tungsten pig
with an assumed matrix material of LiOH 1.6 g cc⁻¹.

Table 4-5. RSS agreement between the C-series sources and the C-series generated
spectrum for different matrices.

<table>
<thead>
<tr>
<th>Matrix, density (g cc⁻¹)</th>
<th>RSS</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiOH, 1.6 (calculated from response function)</td>
<td>1.3*10⁻⁶</td>
</tr>
<tr>
<td>LiOH, 1.32</td>
<td>2.0*10⁻⁵</td>
</tr>
<tr>
<td>Li2O, 0.75</td>
<td>4.8*10⁻⁴</td>
</tr>
<tr>
<td>LiH, 0.56</td>
<td>1.6*10⁻⁵</td>
</tr>
<tr>
<td>LiH, 1.0</td>
<td>1.4*10⁻⁴</td>
</tr>
<tr>
<td>LiOH, 1.6 (calculated directly in MCNP)</td>
<td>1.2*10⁻⁶</td>
</tr>
<tr>
<td>LiOH, 1.6 (MRC encapsulation)</td>
<td>1.8*10⁻⁵</td>
</tr>
<tr>
<td>LiOH, 1.6 (Gammatron encapsulation, no tungsten pig)</td>
<td>2.1*10⁻⁶</td>
</tr>
</tbody>
</table>
The effect of AmO$_2$ particle size was simulated. The minimum and maximum bounds of the lithium spectrum are plotted in Figure 4-7 with the expanded bounds as dashed lines. The spectra were generated by fitting within these bounds to minimize the RSS. The effect on emergent alpha particle energy as the AmO$_2$ radius increases as simulated in MCNP6 is shown in Table 4-6. The corresponding lithium and oxygen average energies as calculated in SOURCES4C are also shown. The asymptotic nature of the Li and O neutron energies demonstrates how reaction energy thresholds interact with the decreasing alpha kinetic energy.
Figure 4-7. Bounds for fitting the AmLi spectra. Photo credit: Weinmann-Smith.

Table 4-6. Alpha and neutron average energies as a function of AmO$_2$ particle size.

<table>
<thead>
<tr>
<th>Particle radius (micrometer)</th>
<th>Emergent average alpha energy (MeV)</th>
<th>Li ($\alpha$,n) average neutron energy (MeV)</th>
<th>O ($\alpha$,n) average neutron energy (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>5.36</td>
<td>0.560</td>
<td>2.43</td>
</tr>
<tr>
<td>1</td>
<td>5.22</td>
<td>0.528</td>
<td>2.39</td>
</tr>
<tr>
<td>3</td>
<td>4.68</td>
<td>0.501</td>
<td>2.29</td>
</tr>
<tr>
<td>5</td>
<td>4.09</td>
<td>0.495</td>
<td>2.25</td>
</tr>
<tr>
<td>7</td>
<td>3.39</td>
<td>0.494</td>
<td>2.24</td>
</tr>
<tr>
<td>9</td>
<td>2.79</td>
<td>0.494</td>
<td>2.24</td>
</tr>
</tbody>
</table>

The generated spectra for the C-series average, N-series average, MRC-series average, and N-437 source are plotted in Figs. 4-8, 4-9, 4-10, and 4-11. The RSS agreements of the spectra with the measurements and the relative lithium contribution by neutron emission rate is given in Table 4-8. N-437 was chosen for an individual spectrum because it was the hardest to fit with the lowest inner ring ratio. The lithium and oxygen energy bounds were expanded to a factor of 5 and the restriction on the adjacent bin rate of change was lifted to allow an RSS agreement to the level of the Cf-
252 sources. These spectra are emergent from the (α,n) interactions and should be modeled as a neutron source in a volume of the AmLi matrix in the source encapsulation.

The rate of change limitation on adjacent bins was removed to demonstrate its effects. The spectra are shown in Figs. 4-12, 4-13, and 4-14 with the agreement and lithium contribution in Table 4-9. The spectra have stronger lithium peaks and a more gradual change between lithium and oxygen. The lithium peak intensity increases with decreasing restrictions, which suggests the optimal mathematical solution is a monoenergetic spectrum at that energy. Both the restricted and unrestricted spectra agree with the measurements as well as the Cf-252 source agreement, but the rate of change restriction limits the unnatural peak feature.

Figure 4-8. AmLi C-series generated spectrum. Photo credit: Weinmann-Smith.
Figure 4-9. AmLi N-series generated spectrum. Photo credit: Weinmann-Smith.

Figure 4-10. AmLi MRC-series generated spectrum. Photo credit: Weinmann-Smith.
Figure 4-11. AmLi N-437 generated spectrum. Photo credit: Weinmann-Smith.

<table>
<thead>
<tr>
<th>Source</th>
<th>RSS</th>
<th>Lithium contribution</th>
</tr>
</thead>
<tbody>
<tr>
<td>C-series</td>
<td>$1.3 \times 10^{-6}$</td>
<td>93.3%</td>
</tr>
<tr>
<td>N-series</td>
<td>$1.4 \times 10^{-6}$</td>
<td>91.9%</td>
</tr>
<tr>
<td>MRC-series</td>
<td>$1.1 \times 10^{-6}$</td>
<td>93.3%</td>
</tr>
<tr>
<td>N-437</td>
<td>$1.4 \times 10^{-6}$</td>
<td>92.1%</td>
</tr>
</tbody>
</table>
Figure 4-12. AmLi N-series spectrum without adjacent bin rate of change restrictions. Photo credit: Weinmann-Smith.

Figure 4-13. AmLi C-series spectrum without adjacent bin rate of change restrictions. Photo credit: Weinmann-Smith.
Figure 4-14. AmLi MRC-series spectrum without adjacent bin rate of change restrictions. Photo credit: Weinmann-Smith.

Table 4-8. RSS agreement and lithium contribution for the unrestricted AmLi spectra.

<table>
<thead>
<tr>
<th>Source</th>
<th>RSS</th>
<th>Lithium contribution</th>
</tr>
</thead>
<tbody>
<tr>
<td>C-series</td>
<td>1.0*10^{-6}</td>
<td>92.8%</td>
</tr>
<tr>
<td>N-series</td>
<td>1.3*10^{-6}</td>
<td>91.8%</td>
</tr>
<tr>
<td>MRC-series</td>
<td>9.3*10^{-7}</td>
<td>92.8%</td>
</tr>
</tbody>
</table>

The effectiveness of the generated spectra in reproducing the AmLi measurements were compared to spectra from the literature. The historical spectra RSS agreements with the C-series, N-series, MRC-series, and N-437 source measurements are shown in Figure 4-15. The best agreements are bolded. The first four spectra are this work. Measured spectra are listed next. They were measured after interaction with the AmLi matrix and encapsulation so these were removed from the simulation. The Geiger and below spectra are calculations of the neutron creation by (α,n) interactions before they interact with the matrix or encapsulation. The last spectrum, SOURCES4C, was generated by simulating the AmLi matrix, LiOH 1.6 g cc^{-1}, in SOURCES4C. In
SOURCES4C a uniform distribution of atoms in an infinite matrix is assumed, there are no particles simulated. The spectrum has the poorest performance, suggesting that this simplification introduces significant errors.

Finally, select spectra were used to simulate a measurement of 2kg of HEU metal in the AWCC. The range of average energies, detection efficiency of the AmLi neutrons, doubles rate, and U-235 mass are shown in Table 4-10. The sensitivity of U-235 mass to choosing the best performing spectra is 3.8%, and 5.6% when including the worst performing spectrum. This demonstrates the significant range in spectra and the need to choose a spectrum based on the source used for measurements in order to produce accurate simulations.

**Discussion**

These measurements have quantified the neutron energy differences between many AmLi sources. In the simulation of measurements it is crucial to choose a spectrum that accurately represents the AmLi source used. There is not one single spectrum that best represents all sources. The many literature spectra are all likely the best representation of the AmLi source that they measured. The spectra generated here are from averages of several source measurements from the same manufacturer. A user with a source from one of these manufacturers will hopefully find these spectra more accurate than choosing their favorite from the literature even if it does not match exactly. For simulations where high precision is needed, the uncertainty introduced by the AmLi spectrum has been quantified, guiding users to determine if they should measure the spectrum of the source they will use for the highest accuracy.
<table>
<thead>
<tr>
<th>Spectrum</th>
<th>C-series RSS</th>
<th>N-series RSS</th>
<th>MRC-series RSS</th>
<th>N-437 source RSS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Generated C-series</td>
<td>$1.3 \times 10^{-6}$</td>
<td>$4.9 \times 10^{-6}$</td>
<td>$5.8 \times 10^{-6}$</td>
<td>$9.5 \times 10^{-5}$</td>
</tr>
<tr>
<td>Generated N-series</td>
<td>$4.8 \times 10^{-5}$</td>
<td>$1.4 \times 10^{-6}$</td>
<td>$3.0 \times 10^{-6}$</td>
<td>$9.9 \times 10^{-6}$</td>
</tr>
<tr>
<td>Generated MRC-series</td>
<td>$5.7 \times 10^{-5}$</td>
<td>$2.6 \times 10^{-6}$</td>
<td>$1.1 \times 10^{-6}$</td>
<td>$6.0 \times 10^{-6}$</td>
</tr>
<tr>
<td>Generated N-437</td>
<td>$9.4 \times 10^{-5}$</td>
<td>$9.4 \times 10^{-6}$</td>
<td>$6.1 \times 10^{-6}$</td>
<td>$1.4 \times 10^{-5}$</td>
</tr>
<tr>
<td>Birch</td>
<td>$3.4 \times 10^{-5}$</td>
<td>$1.3 \times 10^{-4}$</td>
<td>$1.5 \times 10^{-4}$</td>
<td>$2.0 \times 10^{-4}$</td>
</tr>
<tr>
<td>Delafield</td>
<td>$4.7 \times 10^{-5}$</td>
<td>$1.1 \times 10^{-5}$</td>
<td>$1.5 \times 10^{-5}$</td>
<td>$2.5 \times 10^{-5}$</td>
</tr>
<tr>
<td>Ing</td>
<td>$4.9 \times 10^{-5}$</td>
<td>$2.5 \times 10^{-5}$</td>
<td>$3.6 \times 10^{-5}$</td>
<td>$4.9 \times 10^{-5}$</td>
</tr>
<tr>
<td>Obninsk</td>
<td>$2.4 \times 10^{-5}$</td>
<td>$6.0 \times 10^{-5}$</td>
<td>$7.7 \times 10^{-5}$</td>
<td>$1.9 \times 10^{-4}$</td>
</tr>
<tr>
<td>Owen 0.1</td>
<td>$4.2 \times 10^{-4}$</td>
<td>$1.9 \times 10^{-4}$</td>
<td>$1.8 \times 10^{-4}$</td>
<td>$1.3 \times 10^{-4}$</td>
</tr>
<tr>
<td>Owen 5</td>
<td>$6.5 \times 10^{-4}$</td>
<td>$3.6 \times 10^{-4}$</td>
<td>$3.5 \times 10^{-4}$</td>
<td>$2.8 \times 10^{-4}$</td>
</tr>
<tr>
<td>Tagziria 2003</td>
<td>$1.0 \times 10^{-4}$</td>
<td>$1.9 \times 10^{-4}$</td>
<td>$2.2 \times 10^{-4}$</td>
<td>$2.6 \times 10^{-4}$</td>
</tr>
<tr>
<td>Tagziria 2004</td>
<td>$8.3 \times 10^{-4}$</td>
<td>$4.9 \times 10^{-4}$</td>
<td>$4.6 \times 10^{-4}$</td>
<td>$3.7 \times 10^{-4}$</td>
</tr>
<tr>
<td>Werle</td>
<td>$5.3 \times 10^{-5}$</td>
<td>$1.9 \times 10^{-4}$</td>
<td>$2.1 \times 10^{-4}$</td>
<td>$2.8 \times 10^{-4}$</td>
</tr>
<tr>
<td>Geiger</td>
<td>$4.6 \times 10^{-5}$</td>
<td>$2.3 \times 10^{-5}$</td>
<td>$3.2 \times 10^{-5}$</td>
<td>$4.6 \times 10^{-5}$</td>
</tr>
<tr>
<td>Tagziria 2012</td>
<td>$9.5 \times 10^{-5}$</td>
<td>$5.5 \times 10^{-5}$</td>
<td>$3.5 \times 10^{-5}$</td>
<td>$3.5 \times 10^{-5}$</td>
</tr>
<tr>
<td>Sources99</td>
<td>$3.1 \times 10^{-4}$</td>
<td>$1.3 \times 10^{-4}$</td>
<td>$1.3 \times 10^{-4}$</td>
<td>$9.2 \times 10^{-5}$</td>
</tr>
<tr>
<td>SOURCES4C</td>
<td>$1.1 \times 10^{-2}$</td>
<td>$9.4 \times 10^{-3}$</td>
<td>$9.4 \times 10^{-3}$</td>
<td>$9.0 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

Figure 4-15. RSS agreements between measurements and historical spectra and spectra from this work simulated in the 5RMC MCNP6 model. Photo credit: Weinmann-Smith.
Table 4-9. Range of U-235 masses simulated by different spectra.

<table>
<thead>
<tr>
<th>Performance in simulating AmLi measurements</th>
<th>Spectrum</th>
<th>Avg. Energy (keV)</th>
<th>Efficiency</th>
<th>Doubles</th>
<th>Mass (g U-235)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Best (this work)</td>
<td>Generated MRC</td>
<td>547</td>
<td>0.069</td>
<td>135</td>
<td>1947</td>
</tr>
<tr>
<td>Best (measured)</td>
<td>Delafiel</td>
<td>474</td>
<td>0.066</td>
<td>140</td>
<td>2014</td>
</tr>
<tr>
<td>Best (calculated)</td>
<td>Geiger</td>
<td>536</td>
<td>0.070</td>
<td>134</td>
<td>1938</td>
</tr>
<tr>
<td>Worst</td>
<td>Tagziria 2004</td>
<td>556</td>
<td>0.071</td>
<td>132</td>
<td>1903</td>
</tr>
<tr>
<td>Range</td>
<td></td>
<td>82</td>
<td>0.006</td>
<td>8</td>
<td>111</td>
</tr>
</tbody>
</table>

The use of the RSS to quantify agreement simplifies comparison to a single number but is unintuitive in understanding if differences are important or not. Still, there are some clues to help clarify its meaning. The agreement between Cf-252 sources is about $1 \times 10^{-6}$ while AmLi agreement is about $1 \times 10^{-5}$, and the effects of different matrices and encapsulations is calculated. The simulated U-235 masses with different spectra are easier to understand, and the RSS of each spectrum is available.

There are several limitations and assumptions in the generation of the spectra. So many factors in the simulation are unknown: the AmO$_2$ particle size, the AmLi matrix, the degree of mixing, the polyethylene density, and more. The generated spectra are not what is emitted from the AmLi source in the real world. Being limited to 5 measured values, the 5 ring ratios, makes that impossible to find with this experiment, and the difficulties of neutron spectroscopy makes finding the spectrum difficult even with optimized equipment. Instead the spectra are what accurately simulate the measurements in the 5RMC. It is assumed that all of the unknowns average out and are
handled by the fitting. Even if the right matrix or density is not simulated, the final ring ratios and hopefully U-235 fission is correct. In fact, it is known that the 5RMC model is not exactly correct because it did not exactly simulate the Cf-252 measurements. Thus these spectra are most accurate for this 5RMC model and any application to other models should include an adjustment to remove the 5RMC bias and include whatever biases exist in the new models. Still, the improved accuracy from using a manufacturer specific spectrum should overcome these uncertainties.

**Conclusions**

This work comprises three main results. First, for the first time measurements were used to characterize the variations in AmLi and Cf-252 sources. A standard was generated for spectral fitting quality based on the Cf-252 variation. Second, simulations and modeling were used to generate spectra specific to Gammatron N, Gammatron C, and MRC-series of sources and individual sources. These spectra agree with the measurements to within the variation between Cf-252 sources. Using a series-specific spectrum provides researchers with improved precision in their simulation of AmLi sources, reducing the spectrum-dependent systematic uncertainty. Finally, the sensitivity of many aspects of simulating AmLi sources were studied. These included the choice of AmLi spectra including all known published spectra and their effect on U-235 mass. The variation between AmLi sources demonstrated the importance of choosing a spectrum specific to the source used as a spectrum accurately represents the source it was made from but will not represent other sources. Further, the limited set of measurements in this work suggest an even larger variation in AmLi sources around the world.
While this was the most comprehensive study of the variation in AmLi spectra to date, the comparison metric was limited and improved practical application was not demonstrated in a specific assay system. A detector like the Uranium Neutron Coincidence Collar should be used to understand the improvement in accuracy of assay of U-235 mass. The system should be calibrated through simulations with a specific source’s spectrum and confirmed with measurements using the same source. This would quantify the improved accuracy of the spectrum, and, if accurate enough, demonstrate the new and valuable capability of simulated calibration measurements.
CHAPTER 5
A COMPARISON OF FISSION MODELS

Introduction

Monte Carlo simulations are often used in the design and optimization of neutron multiplicity counters for international safeguards. The simulations typically reach an agreement with measurements of about 5%-10%, depending on the application. The 5%-10% agreement is systematic uncertainty, the simulations and measurements are both run long enough for statistical uncertainty to be negligible in the design and optimization phase. The systematic uncertainty comes from discrepancies in the physical description of the detector, uncertainties in cross sections for particle interaction and transport, and uncertainties in the radiation source information. The radiation source information includes the neutron emission rate, which is well known for Cf-252 but has large uncertainties for (α,n) sources, neutron energy spectra which is poorly known for the less common interactions and isotopes, the multiplicity distributions which are generally well known, and correlations between fission particles which are currently being investigated with new models being developed.

The desire to model these correlations has motivated the development of the Fission Reaction Event Yield Algorithm (FREYA) and Cascading Gamma-Ray Multiplicity with Fission (CGMF). FREYA was developed at Lawrence Livermore National Lab and CGMF was developed at Los Alamos. Both model individual fission events with detailed physics to generate the fission products and emitted radiation. This is in contrast to the capabilities of the most recent MCNP release, MCNP6.1.1b, where

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the properties of fission radiation are sampled independently from each other from lookup data, the fission itself is not simulated, and there are no correlations except for the number of neutrons emitted, or multiplicity. Because the correlations of particles emitted in fission events are valuable observables used in radiation detectors including multiplicity counters, accurately simulating fission events is needed. FREYA and CGMF have been incorporated into MCNP6.2, to be released late 2018, to address this issue. This work compared the use of FREYA, CGMF, and another correlated model, MCNP-PoliMi, to the standard simulations of MCNP6.1.1b in modeling common safeguards detectors. The performance of each model was evaluated and a recommendation was made to guide users on the choice of model upon the release of MCNP6.2. Since MCNP6.2 has not been released yet, the CGMF and FREYA results were generated with MCNP6.20 prerelease ID 06/09/17.

FREYA and CGMF simulate individual analog fissions, conserving momentum, energy, and angular momentum [64, 65]. The compound nucleus, scission into fission fragments, and deexcitation into fission products are all simulated in detail. The resulting neutrons and photons are correlated from this process which is a remarkable advancement in Monte Carlo simulation because it includes all correlations that would be generated by our current understanding of fission, even if the correlations are not currently known. The resulting neutron energies and multiplicities are also generated completely by the modeling of the fission process, they are not looked up from tables or formulas as in the standard model.

The most significant differences in the models are the neutron energies, multiplicities, and angular correlations, and each was isolated and studied. The angular
correlations are a new addition to MCNP6.2 and were shown to have the greatest effect. The differences in energy distribution had the second greatest effect, and are poorly known so comparison to a ‘true’ measured value is often impossible. Differences in multiplicity had the smallest effect and the multiplicity is the most well-known from measurements. The additional coupled effects are difficult to isolate in the simulation, and so they are included but are expected to have a small impact. So far the models have accurately simulated available measurements [66]. More complex correlations are included. The angular correlation is more extreme at higher neutron energies and lower multiplicities [67, 68]. The neutron multiplicity and energy are correlated [68, 69]. The photon multiplicity and neutron multiplicity are negatively correlated [69]. Note that these correlations are only relevant for low multiplication systems. At high multiplications the correlations average out through multiple events in a fission chain and can no longer be detected. Other work has investigated the fission models for high multiplication systems, while low multiplication is more relevant to safeguards [70].

PoliMi is a sort of hybrid with only some correlations and was designed to more accurately simulate radiation measurements [71]. It was developed for MCNPX and greatly improved the accuracy of that code at the time, but has started to be surpassed by the inclusion of models like FREYA and CGMF. In PoliMi the spontaneous fissions are modeled, the neutron spectrum depends on multiplicity, and the neutrons have an angular correlation [72]. For induced fission an option is given for the source of multiplicity data, and in this work the measurements of Holden and Zucker was used [73]. The PoliMi simulations of this work were performed with the RSICC release version of MCNPX-POLIMI.
A typical MCNP6.1.1b multiplicity counter simulation would use FMULT method=3 data=3 shift=1 to control the generation of fission neutrons [31]. This is referred to as the 'standard model' in this work and is the point of comparison for CGMF, FREYA, and PoliMi. In the standard model, neutrons from induced fission are handled by taking the average neutron multiplicity $\bar{v}$ from the Evaluated Nuclear Data File (ENDF) at the incident neutron energy. The multiplicity distribution is then generated by sampling from a Gaussian distribution that preserves $\bar{v}$. This distribution is adjusted to prevent the sampling of negative neutrons while preserving $\bar{v}$.

Spontaneous fission $\bar{v}$ and multiplicity distributions come from measured data as referenced in the MCNP manual. Neutron energies are chosen from Watt spectra. The Watt spectra parameters used for induced fission are taken from the data library and for spontaneous fission are given in the manual, which were generated from the Madland-Nix model [74]. The standard model simulations were performed with the same code as FREYA and CGMF, MCNP6.20 prerelease ID 06/09/17.

The isotopes most often measured in multiplicity counters are Cf-252, U-235, U-238, Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242, so they were the focus of this work. The spontaneous fission of the even isotopes and the induced fission of the odd isotopes were studied. FREYA and the standard model have data for all of these isotopes. PoliMi is missing data for Pu-238, which is the most uncommon isotope. CGMF is missing data for Pu-238 and U-238, restricting its application in uranium fuel assembly self-interrogation which is driven by U-238 spontaneous fission. When CGMF is called to simulate these isotopes MCNP instead uses the Lawrence Livermore National Laboratory (LLNL) fission model.
The choice of fission model is especially important in simulating Cf-252 interrogation, so that is the application of focus for this work. AmLi sources have been preferred for active interrogation of uranium because the low energy neutrons limit U-238 induced fission. However, reliable AmLi sources are no longer being produced and so Cf-252 is being investigated as a replacement. Cf-252 interrogation sources have been studied in the Active Well Coincidence Counter and the Uranium Neutron Coincidence Collar and are also incorporated in the Advanced Experimental Fuel Counter (AEFC) [75, 76, 77]. Neutrons emitted from Cf-252 have a fission energy spectrum and so they are more likely to induce U-238 fission compared to AmLi. Since AmLi is an \((\alpha,n)\) source the emitted neutrons are random and do not contribute to real coincidence counts, so its contribution is easily accounted for. Cf-252 on the other hand is a spontaneous fission source which does emit coincidence neutrons. As one of these neutrons induces fission, the produced induced fission neutrons are time correlated to the remaining Cf-252 spontaneous fission neutrons. Thus the event multiplicity is the sum of the induced fission and spontaneous fission multiplicities minus the captured neutron. The desired observable, induced fission, is coupled to the spontaneous fission. Thus the treatment of any correlations in the spontaneous fission is especially important to accurately modeling the whole event. For example, angular correlations must be included because if neutrons are more likely to be emitted in the same direction it is more likely that two induced fissions will occur from the same spontaneous fission. If neutrons are more likely to be emitted at 180 degrees it is more likely that only one induced fission will occur per spontaneous fission, because if a neutron is emitted towards the sample another neutron will be emitted away from it. These correlations are
especially important for interrogation sources because they are rarely placed in the center of the detector, unlike the assay sources.

The purpose of this work is to guide users on the choice of fission model for their application. In some cases one model is clearly more accurate than the others. Sometimes the models are significantly different but measurements are inadequate to judge which result is closest to the ‘true’ value. The major differences between the models are the energies, multiplicities, and angular correlations. First, the models’ production of these values is compared to each other and to measurement consensus when available. Then, each difference was studied independently through gedanken simulations which isolated a single effect. These simulations demonstrated the magnitude or importance of each effect. For example, to study differences in multiplicity distribution, a point source in a thick 4pi detector was simulated with each model. The results also indicated the sensitivity of different detector designs, for example the importance of energy spectra at different detector thicknesses. Then the models were evaluated in the simulation of measurements taken with the AWCC and AEFC. Finally, PTRAC results calculated the probabilities of different correlated events which lead to the different AWCC results for each model.

**Simulation and measurements**

The first simulation was to compare the run times for a typical safeguards detector. The AWCC was simulated for Cf-252 interrogation of uranium. The run times affect the practicality of using the models.

The emitted neutron properties for spontaneous and induced fission were simulated. The values were taken from the MCNP output file except for the angular correlation which was calculated with PTRAC. The average neutron multiplicity was
found for each model and isotope. Spontaneous fission results were compared to the measured consensus values reported in Santi and Miller [78]. Induced fission results were compared to measured values from Holden and Zucker [73]. Again, the incident thermal neutron energy for induced fission was 0.253 eV.

The average neutron energy was found. Measured data was not adequate for comparison except for Cf-252, which is well known to be $2.122 \pm 0.017$ MeV [79, 80].

The neutron-neutron angular correlation was found through PTRAC. The cosine of the angle between two neutrons was found such that neutrons moving in opposite directions at 180 degrees have a cosine of -1. The cosine between every pair of neutrons emitted in a single fission was recorded, so a fission that emitted $n$ neutrons has $n^*(n-1)/2$ pairs. In PoliMi source events the PoliMi model is called after the source event is written to PTRAC, so the PTRAC source events are incorrect. This will not affect the angular correlation simulations because the neutron direction cosine was recorded as the particles cross a surface after PoliMi has rewritten their starting source information.

The multiplicity distributions were recorded and compared to the same measured consensus values as the multiplicity averages. The energy distributions were recorded. The Cf-252 energy distribution was compared to the spectrum of ENDF/B-VIII which is taken from Manhart [81], a measurement consensus evaluation with uncertainties less than 2% in the range of 180 keV to 9.3 MeV. The Cf-252 comparison is in terms of standard deviations away from the Manhart spectrum.

After the neutron properties were calculated, gedanken simulations were ran. The simulations were a thought experiment designed to isolate the multiplicity
distributions, angular correlations, and energy distributions. The simulations were of a Cf-252 point source in the center of a detection sphere. This approximates the geometry of a well counter like the AWCC. The sphere consisted of a homogeneous mixture of moderating polyethylene and absorbing He-3. Neutron coincidence captures on the He-3 were tallied with a typical detector coincidence gate structure of 1.5 microsecond predelay and 64 microsecond gate width. The doubles rate was used as the comparison metric. The results are reported in percent difference from the standard model for each of FREYA, CGMF, and PoliMi, for ease of comparison and to see the effect of changing from the standard model to each of the other models. The sphere fully enclosed the point source to remove dependency on angular correlation. It was thick to remove dependency on neutron energy. The standard model neutron multiplicity could be set to the same multiplicity as the FREYA, CGMF, or PoliMi models to remove dependency on the multiplicity distribution.

First, the point source was simulated with the different fission models in a 45 cm thick, 4pi detector and the doubles rate was recorded to show the effects of the different multiplicity distributions.

Then, the standard model was forced to have each of the other model’s multiplicity distribution to remove that effect. The detector solid angle was changed by removing ‘orange slices’ from the sphere. In other words the sphere azimuthal was increased in 60 degree increments to cover solid angles of 0.67, 1.33, 2, 2.67, 3.33, and 4pi. These cases are shown in Figure 5-1.
Finally, with the multiplicities fixed and the solid angle at $4\pi$, the detector thickness was changed from 0 to 45cm to study the effect of different neutron energies and distributions on the detected doubles rate.

![Figure 5-1](image1.png)

**Figure 5-1.** Sphere detector with ‘orange slice’ cut to vary solid angle with Cf-252 point source at the center. Solid angles of $2\pi$ (left) and $3.33\pi$ (right) are shown. Photo credit: Weinmann-Smith.

A second gedanken experiment was chosen to better apply to passive self-interrogation of fuel assemblies in the UNCL. This simulation consisted of a point source of $^{238}\text{U}$ spontaneous fission in a cylinder of polyethylene and He-3 mixture. The cylinder thickness was changed to study the effect of $^{238}\text{U}$ energy and the cylinder height was changed to study the effects on angular correlation for a cylinder geometry, Figure 5-2.

![Figure 5-2](image2.png)

**Figure 5-2.** Simulation of point source of $^{238}\text{U}$ spontaneous fission in a cylinder of polyethylene and He-3 mixture.

An experimental campaign was carried out as part of other work [76, 75]. The fission models were used to simulate the measurements for comparison. Cf-252 was measured in the AEFC fuel through-hole, Figure 5-3, in the center and raised and lowered by 5cm. The AWCC was used for the second set of measurements. The UISO cans of uranium oxide were placed on the bottom of the sample cavity. The plug rings were removed as shown in Figure 5-4, and the cavity height was 20 cm. The UISO cans
are thin walled with U$_3$O$_8$ powder at about 2.3 g/cc density. Each can contains 1 kg of uranium with enrichments of 17%, 38%, 52%, 66%, and 91%. Both AmLi and Cf-252 were used for the AWCC interrogation source. Only a bottom plug source was used, the top plug was empty. For the AWCC UISO-17 case the doubles rates were converted to mass using an AWCC calibration curve constructed from the UISO measurements. The result shows the mass effect of the fission models, but note that actual AWCC calibration measurements would be conducted very differently.

Figure 5-2. Cylinder gedanken experiment to study the UNCL effects. Photo credit: Weinmann-Smith.

Figure 5-3. AEFC system. Photo credit: Menlove [77].
Finally, an analysis of the MCNP PTRAC files was performed to tally additional aspects of the simulation. PTRAC files are a record of the events in the simulation, and can include source events, bank events, surface crossing events, collision events, and termination events. Analyzing the PTRAC file allows a user to essentially make their own tallies which can use more complex logic than is available by default in MCNP. All of the PTRAC results are normalized to the number of spontaneous fissions, and are reported as the probability of an event occurring per spontaneous fission. The goal was to better understand why FREYA reported lower results which were closer to measurements, and to better understand the effects of the Cf-252 ‘boost’ to the doubles rate. The PTRAC analysis found the number of induced fissions for each of the models. The probability of a given number of induced fissions from a single spontaneous fission was found to search for large fission events which would potentially bias results. Then, the number of spontaneous fission or induced fission neutrons detected in the He-3 tubes was found. Finally, the number of histories with both a Cf-252 and uranium
neutron detection were found. This showed whether the new angular correlations of FREYA and CGMF significantly impacted the agreement with measurements.

**Results**

A typical simulation of the AWCC took the following run times relative to the standard model, Table 5-1. Note PoliMi's increased speed due to running on the faster MCNPX.

<table>
<thead>
<tr>
<th></th>
<th>Standard</th>
<th>FREYA</th>
<th>CGMF</th>
<th>PoliMi</th>
</tr>
</thead>
<tbody>
<tr>
<td>Relative time</td>
<td>1.0</td>
<td>1.4</td>
<td>1250</td>
<td>0.9</td>
</tr>
</tbody>
</table>

The average neutron multiplicity for spontaneous and induced fission is shown in Figure 5-5. The published results are from Santi and Miller for spontaneous fission and Holden and Zucker for thermal neutron induced fission. Table 5-2 describes the standard deviations away from the measured values for the 4 isotope-model multiplicities that differ from measurements by more than 3 standard deviations. FREYA has significant deviations for Pu-240 which contributes the most spontaneous fission neutrons for plutonium measurements.

The average fission neutron energy is shown in Figure 5-6. There are significant variations greater than 150 keV for Cf-252, Pu-239, and Pu-240. FREYA's average Cf-252 energy is 2.283 MeV which is greater than the accepted Cf-252 average energy by 150 keV.

Angular correlations, or angle between neutron pairs, is shown in Figure 5-7. The kinematic boost from the momentum of the fission fragments causes an increase in neutrons emitted in the same and opposite directions for FREYA and CGMF, while
neutrons emitted at 90 degrees are least likely. The opposite direction is more likely than the same direction because the same direction can only be favored when a disproportionate number of neutrons are emitted from the same fragment [68]. Every angle between pairs is equally likely in the standard model because the neutrons are emitted independent from each other. PoliMi is most likely to simulate neutrons in the same direction, and this effect is much stronger than in FREYA and CGMF. The neutron energy is sampled from the Watt distribution which is used to calculate the neutron angle relative to the fission fragment direction.

![Average neutron multiplicity of spontaneous and induced fission for the fission models of MCNP compared to values from Santi and Miller and Holden and Zucker. Photo credit: Weinmann-Smith.](Figure 5-5)

Figure 5-5. Average neutron multiplicity of spontaneous and induced fission for the fission models of MCNP compared to values from Santi and Miller and Holden and Zucker. Photo credit: Weinmann-Smith.

The multiplicity distributions for the various isotopes are shown in Figs. 5-8 through 5-15. FREYA and CGMF can simulate large multiplicities up to 11+ while PoliMi and the standard model are limited by available data which has multiplicities to 5-6 for all but Cf-252. For U-238 FREYA for example, multiplicities 5 through 11 consist of
0.44% of the multiplicities, so the importance of the higher multiplicities may be negligible for coincidence counting. This extended range shows the advantage of using models. However, models also reduce accuracy. For the most probable multiplicities the models disagree with measured values by as many as 30 standard deviations, for example in Pu-240, Figure 5-13. For spontaneous fission the standard model and PoliMi use the measured data so they agree with it exactly. Note that for the Pu-238 and U-238 results CGMF uses the LLNL model because it does not have data for those isotopes. There is no clear trend for induced fission. For U-235 CGMF and the standard model disagree with measurements at most multiplicities, while the other models still disagree at two multiplicities. For Pu-239 FREYA and the standard model disagree with measurements on multiplicities 0 and 2. PoliMi exactly matches each multiplicity because it uses the measurement data directly.

Table 5-2. Number of standard deviations away from measured average multiplicities.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Std. Dev. From measured</th>
<th>Model</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-240</td>
<td>17</td>
<td>FREYA</td>
</tr>
<tr>
<td>Pu-241</td>
<td>6</td>
<td>FREYA</td>
</tr>
<tr>
<td>U-235</td>
<td>3</td>
<td>Standard</td>
</tr>
<tr>
<td>U-235</td>
<td>3</td>
<td>PoliMi</td>
</tr>
</tbody>
</table>

The energy distributions for the various isotopes are shown in Figs. 5-16 and 5-19 through 5-25. The energy distribution can differ even when the average neutron energy is exactly the same, as in the U-235 energies for FREYA and CGMF which are both 1.94 MeV. Previous work has shown that this difference can cause significant changes in the detection efficiency and thus coincidence rate due to the detector's
complex response functions [82]. For the isotopes other than Cf-252, PoliMi and the standard model have identical energy distributions and average energies. FREYA always has a stronger peak at around 750 keV. Cf-252 is shown in log scale at high energies, Figure 5-16 insert, to show the high energy tail that causes FREYA to have such a high average energy compared to the accepted value. The Manhart evaluated measured consensus Cf-252 spectrum is shown in Figure 5-17 along with the model spectra for a reduced range to show detail in the plot. The number of standard deviations away from the Manhart spectrum for the energy range with good agreement, 0.2 to 9.3 MeV, is shown in Figure 5-18. The results show clear divergence at energies above 5 MeV for CGMF and especially for FREYA.

![Average fission neutron energy of spontaneous and induced fission for the fission models of MCNP. Photo credit: Weinmann-Smith.](image)

Figure 5-6.
Figure 5-7. Angle between neutron pairs for the various fission models. Photo credit: Weinmann-Smith.

Figure 5-8. Cf-252 SF multiplicity distribution. Photo credit: Weinmann-Smith.
Figure 5-9. U-235 IF multiplicity distribution. Photo credit: Weinmann-Smith.

Figure 5-10. U-238 SF multiplicity distribution. Photo credit: Weinmann-Smith.
Figure 5-11. Pu-238 SF multiplicity distribution. Photo credit: Weinmann-Smith.

Figure 5-12. Pu-239 IF multiplicity distribution. Photo credit: Weinmann-Smith.
Figure 5-13. Pu-240 SF Multiplicity distribution. Photo credit: Weinmann-Smith.

Figure 5-14. Pu-241 IF multiplicity distribution. Photo credit: Weinmann-Smith.
Figure 5-15. Pu-242 SF multiplicity distribution. Photo credit: Weinmann-Smith.

Figure 5-16. Cf-252 SF energy distribution. Photo credit: Weinmann-Smith.
Figure 5-17. Cf-252 SF energy distribution compared to Manhart. Photo credit: Weinmann-Smith.

Figure 5-18. Cf-252 SF energy distribution standard deviations away from Manhart. Photo credit: Weinmann-Smith.
Figure 5-19. U-235 IF energy distribution. Photo credit: Weinmann-Smith.

Figure 5-20. U-238 SF energy distribution. Photo credit: Weinmann-Smith.
Figure 5-21. Pu-238 SF energy distribution. Photo credit: Weinmann-Smith.

Figure 5-22. Pu-239 energy distribution. Photo credit: Weinmann-Smith.
Figure 5-23. Pu-240 energy distribution. Photo credit: Weinmann-Smith.

Figure 5-24. Pu-241 energy distribution. Photo credit: Weinmann-Smith.
The gedanken simulation results follow. The effects of the multiplicity distribution were found by simulating a thick 4pi sphere around a Cf-252 point source with the different models, which are shown in Table 5-3. The difference of each model with the standard model is shown. Switching to CGMF is shown to increase the doubles rate by 1.8%, which is attributed entirely to the difference in multiplicity. This is the largest difference and yet is a relatively small effect. The difference between, for example, FREYA and the standard model forced to have FREYA multiplicities is also shown to be 0 within the statistical uncertainty to show that only the multiplicities cause the reported difference. Note that the difference between PoliMi and the standard model is 0 because they both use the Santi multiplicities for Cf-252.

Figure 5-25. Pu-242 energy distribution. Photo credit: Weinmann-Smith.
Table 5-3. Effects of multiplicity distribution on the various models.

<table>
<thead>
<tr>
<th></th>
<th>FREYA</th>
<th>CGMF</th>
<th>PoliMi</th>
</tr>
</thead>
<tbody>
<tr>
<td>Difference with the standard model (%)</td>
<td>0.6 (0.1)</td>
<td>1.8 (0.1)</td>
<td>0.0 (0.1)</td>
</tr>
<tr>
<td>Difference when the standard model has other model multiplicities (%)</td>
<td>0.0 (0.1)</td>
<td>0.1 (0.1)</td>
<td>0.0 (0.1)</td>
</tr>
</tbody>
</table>

The difference in doubles as a function of solid angle is shown in Figure 5-26. ‘Orange slices’ were removed from the sphere to reduce the solid angle. The results are shown as a percent difference from the standard model so the results could be compared graphically. The results are explained through Figure 5-7, the neutron angular correlations. PoliMi is shown to greatly increase the doubles at low solid angles because it has a greatly increased probability of simulating multiple neutrons in the same direction. A double count requires two neutrons to be detected which, at low solid angles, can only happen when they were emitted in the same direction. Compared to the standard model, FREYA and CGMF both are more likely to simulate neutrons in the same direction and so their doubles is also higher at low solid angles. However, they are also more likely to predict neutrons emitted at 180 degrees. As the solid angle increases from 0-2pi, their predicted doubles rate from 180 degree neutrons stays the same while the standard model, which predicts more neutrons emitted from 0-180 degrees, increases. As the solid angle increases from 2-4pi FREYA and CGMF see an increased doubles rate from 180 degree neutrons and the difference between their results and the standard model converges to 0%. The standard model had the other model’s multiplicities so the results are fully attributed to the angular correlations.
Figure 5-26. The change in doubles rate when switching from standard to FREYA, CGMF, and PoliMi models as a function of solid angle for a Cf-252 point source in a thick spherical detector. Photo credit: Weinmann-Smith.

The effect of neutron energy is shown in Figure 5-27 by changing the radial thickness of a spherical 4pi detector. The different multiplicities were accounted for. As the thickness increases more neutrons are detected and the energy differences have a smaller effect. FREYA’s high average energy decreases the doubles rate at low thicknesses as neutrons are more likely to escape the detector. Note that the shape of the energy distribution also plays an important role. PoliMi has a higher average energy than CGMF, but also a higher doubles rate at low thicknesses. While a real detector does not have homogeneous He-3 distribution and is not spherical, some comparison can be made. The radial thickness of the HLNCC-II is 8cm, the UNCL-II (PWR) is 10cm, and the ENMC outer ring is 17cm. Again the differences converge to 0 as the detector approaches a thick, 4pi sphere.
The difference in doubles rate as a function of detector height is shown for the cylindrical detector in Figure 5-28. The cylinder was thick to remove dependence on energy. CGMF is not included in the comparison because it lacks U-238 data. PoliMi and FREYA cause a large increase in the doubles rates at low heights. The standard model is most likely to emit neutrons at 90 degrees where one will escape from the top or bottom of the detector and not make a double. As the detector height increases these neutrons are more likely to be detected, while the detector height has a minimal effect on the detection rate of 180 degree and 0 degree neutron pairs, more likely in the FREYA and PoliMi models. Thus as the detector height increases the differences converge at 0%.

Figure 5-27. The change in doubles rate when switching from standard to FREYA, CGMF, and PoliMi models as a function of detector thickness for a Cf-252 point source in a 4pi spherical detector. Photo credit: Weinmann-Smith.
Figure 5-28. Differences in the doubles rates for a cylindrical detector as a function of detector height. Photo credit: Weinmann-Smith.

Experimental measurements were made with the AWCC and AEFC. The 1-sigma statistical uncertainties from both measurements and simulation are plotted in the figures and are negligible. The measurement results also include the NIST calibrated 1.0% 1-sigma source strength uncertainty.

The first results are the AWCC with an AmLi source interrogating uranium oxide samples, Figure 5-29. There is no statistical difference between the fission models in the AmLi case, and they agree with the measurements. In the AmLi case the fission models only handle induced fission, the AmLi source neutrons are generated randomly. This shows that the differences between the fission models are negligible for induced fission of U-235, which is the dominant driver of the doubles rate. The near-4pi geometry of the source sample may also explain the lack of differences.
Switching the AmLi source for Cf-252 shows a significant difference, Figure 5-30. The models are statistically different from each other and all are far from the measurement results. This shows that the accurate simulation of Cf-252 is crucial for accurate measurement of neutron safeguards systems. PoliMi and CGMF simulate more doubles than the standard model, while FREYA reports less. FREYA’s lower doubles rate is likely due to its higher average Cf-252 neutron energy, the effects of which are shown in the previous results.

The simulation of AEFC measurements is shown in Figure 5-31. The fission models give significantly different results from each other and the measurements. Again FREYA simulates a lower doubles rate because of its higher average Cf-252 neutron energy. While this brings it closer to the measurements, it does not mean the results are more accurate. The CGMF result closely matches the standard model result, which shows that the inclusion of angular correlations is not the key to good agreements with this measurement configuration. However, the AEFC has a small solid angle from the source to the detection He-3 tubes. PoliMi has been shown to simulate an increased doubles rate for low solid angles due to its angular correlation, and that result is confirmed here with the AEFC while worsening measurement agreement.
Figure 5-29. Measured and simulated doubles of an AmLi interrogation source in the AWCC. Photo credit: Weinmann-Smith.

Figure 5-30. Measured and simulated doubles of a Cf-252 interrogation source in the AWCC. Photo credit: Weinmann-Smith.
The UISO measurement results were used to generate a calibration curve for the AWCC, Figure 5-32. Then, the simulations of the UISO-17 case were plotted on the calibration curve. The range of U-235 masses from the different models was 48g. The closest model was FREYA with 241g, and the greatest uncertainty was CGMF with +5g. The measurement result (without the NIST 1% source activity strength uncertainty) gave 176g +4g, which demonstrates how accurate a typical measurement is and how inaccurate the simulations are. This demonstrates why simulations are typically scaled to measurement results and not trusted outright. However, these results are for illustrative purposes only. The range of uranium masses for these UISO samples is too large for the AWCC thermal mode. The samples are thicker than the infinite thickness of the thermal neutrons generated in the AWCC, which can be seen in the UISO-52 and UISO-66 measurements giving almost identical results. To accurately assay these
samples the AWCC should instead be configured with a cadmium liner in fast mode. This was not done because calibration was not the focus of this study. The results are given for the UISO-17 source where this effect is hopefully minimized, but the entire calibration is affected. The result is a curve with a flat slope such that a small change in doubles equates to a large change in uranium mass. Similarly, a small doubles uncertainty results in a large mass uncertainty compared to a steeper slope. The y-axis of the plot starts at the calibration curve y-intercept, 1787. The curve is only valid for the mass range with which it was generated. Unlike with an AmLi curve, the intercept is not 0 because the Cf-252 source causes real coincidences even when the uranium mass is 0g.

Figure 5-32. AWCC calibration curve made with the UISO measurements. Photo credit: Weinmann-Smith.
Finally, analysis of PTRAC outputs was used to calculate some coupled probabilities in the simulation of the UISO-52 measurement to explain why the models give different doubles results. The explanation of these results focuses on FREYA because it most closely matched the measurements. The number of fissions induced per spontaneous fission was calculated. The probability of each number of induced fissions is shown in Figure 5-33. The results show FREYA is slightly less likely to induce every number of fissions from a single Cf-252 spontaneous fission. The total number of induced fissions per spontaneous fission is given in Table 5-4. The number of neutrons from spontaneous fission of Cf-252 or induced fission is also shown in the table. Finally, probability of a Cf-252 neutron being detected given that a uranium neutron has already been detected is shown to quantify the 'boost' to the doubles of the Cf-252. Compared to the standard model, FREYA is 2.0% less likely to simulate an induced fission and 2.3% less likely to simulate detecting a neutron from induced fission. This indicates that induced fission neutrons have a smaller effect on the doubles than the standard model. FREYA simulates 0.3% fewer detections of neutrons from Cf-252. Finally, the probability of detecting a Cf-252 neutron if a uranium neutron is also detected is 0.3% higher for FREYA. The other reduced detection probabilities are the dominant effect that lowers the simulated doubles rate compared to the standard model. The lower induced fission rate and Cf-252 neutron detection rate especially suggest that this is due to the higher average FREYA neutron energy, which leads to fewer interactions. The FREYA model also has a lower U-235 IF $\overline{\nu}$ and a slightly lower U-235 IF average neutron energy.
Figure 5-33. Probability of inducing a given number of fissions per Cf-252 spontaneous fission. Photo credit: Weinmann-Smith.

Table 5-4. Probability of various events related to the doubles rate.

<table>
<thead>
<tr>
<th>Event</th>
<th>Standard</th>
<th>FREYA</th>
<th>CGMF</th>
<th>PoliMi</th>
</tr>
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<tbody>
<tr>
<td>Induced fissions (SF$^{-1}$)</td>
<td>0.45714(5)</td>
<td>0.44805(7)</td>
<td>0.4535(8)</td>
<td>0.44159(9)</td>
</tr>
<tr>
<td>Cf SF neutrons detected (SF$^{-1}$)</td>
<td>0.7300(3)</td>
<td>0.7279(2)</td>
<td>0.7313(3)</td>
<td>0.7316(2)</td>
</tr>
<tr>
<td>U IF neutrons detected (SF$^{-1}$)</td>
<td>0.3281(3)</td>
<td>0.3206(3)</td>
<td>0.3266(6)</td>
<td>0.3266(6)</td>
</tr>
<tr>
<td>Cf neutrons detected given U neutrons detected</td>
<td>0.4854(3)</td>
<td>0.4867(3)</td>
<td>0.4900(8)</td>
<td>0.4900(8)</td>
</tr>
</tbody>
</table>

Discussion

In choosing a fission model for the simulation of thermal neutron detectors there is no obvious choice. This work has demonstrated that every model has limitations. FREYA and CGMF are new and are still undergoing active development so they are
most likely to improve on their worst qualities and take the lead as the best model. However, their nature of calculating everything from fission physics may restrict their progress. While they, especially FREYA, have ‘tweakable’ parameters that are adjusted to match existing data, their model has to generate correct answers for all isotopes, which may not be possible with our existing knowledge of fission physics. Improvements require better measurements and understanding of fission properties. Improving the standard model on the other hand is as simple as programming in the ‘correct’ measurement based data to be used. While this path is much easier and more direct, funding and motivation will likely be focused on the fission models instead. Also, measurement data is not always available, and for many isotopes and energy ranges is severely lacking, such that models are required to fill in the gaps. For example, the standard model’s induced fission multiplicity treatment by creating a Gaussian to preserve \( \nu \) is unphysical and such a fundamental property should be reproduced accurately, but measured multiplicity distributions for a range of energies often do not exist. From a detector designer’s perspective the ideal model is one that reproduces fundamental and full system measurements exactly. Accurately modeling the fundamental properties should result in complex systems being modeled accurately. Modeling the fundamental properties is easily done once they are known, but so much is unknown and better measurements are needed. Even if they are not plugged into a ‘dumb’ model that reproduces them, better measurements are needed to validate the fission models. From this work it remains clear that MCNP simulations will not match measurements until additional research is done.
This work has also shown that the impact of an effect depends on the detector. For example, angular effects do not matter in a 4pi detector. If the standard model has the best energy and multiplicity distribution for the isotope of interest and the detector is 4pi then the standard model should be used instead of FREYA. For a short cylindrical detector the use of FREYA is almost mandatory because the standard model under predicts the doubles by about 10%. However, switching between models in different applications makes the comparison of results meaningless and so this strategy is not suggested, and one model that does everything correct is necessary.

In the broader goal of improved MCNP simulations, accurately reproducing fundamental fission neutron properties is only one part. The system physical description and cross section data must also be accurate. Since the measured result is known, one of these three characteristics can be unknown and solved for to match the measurements. However, currently, all 3 are poorly known such that this method is not possible. The physical descriptions are often lacking because there is no reason to exert the effort for extreme detail when the uncertainties of the other two are so large. The cross sections are also poor relative to other materials because thermal neutron interactions and \( s(\alpha,\beta) \) data for polyethylene is a small niche compared to the industries that drive nuclear data. Further, resolving cross sections or neutron data is a massive undertaking that can only be completed by concentrated effort by a large group of experts and facilities over time. Thankfully this is being done for the fission neutron data, and the physical descriptions can be greatly improved by an individual. There is potential in the next few years to solve the largest unknowns in the modeling of these systems. Then there will be increased motivation and a clearer path forward to solve the
remaining unknowns. While they can be measured directly, there is also potential for a systematic study of where and when measurements do not agree with simulations to identify and reduce the final dominating uncertainties. Note that ‘known’ and ‘unknown’ are relative terms, but the goal should be to reduce the current 10% uncertainties and disagreement with measurements to 1% or better.

Returning to the details of this work’s results, note that the difference between simulation speeds will vary from problem to problem. The simulation of fission events is only one part of the full simulation, and so simulations with more or less other aspects will have different dependencies on the fission model simulation time. The result was of a typical detector, but when simulating, for example, only a point source in a vacuum, the simulation will be almost entirely the fission models and the differences will be exaggerated.

When considering the angular correlation results note that they are a point source in a vacuum. The results are what is emitted out of the atom before any interaction with materials. Others have published experimental measurements and simulations of these experiments measuring this property and the plots have the same axes but different shapes. The measurements are often performed with fast neutron scintillators which are susceptible to cross talk, where a neutron deposits energy in two nearby detectors. This increases the probability of small angle neutron pairs, neutrons in the same direction. The results of this work should not be compared to those plots and the opposite shape, where the 0 degree angle is greater than the 180 degree angle, is expected.
Conclusions

The simulation of individual fission events represents a significant improvement to MCNP capabilities by incorporating fission signatures which were previously unused. This offers new possibilities in radiation detection techniques. However, these events must be shown to correctly reproduce the fundamental fission signatures already relied on for existing neutron detectors for international safeguards. In the comparison of FREYA, CGMF, and PoliMi, to the standard model traditionally used and to measurements of neutron properties and full safeguards detectors, no model clearly outperformed the others. Each model had some limitations, and they are undergoing active development so these results will not always be true. In some areas the best model could not be evaluated because of a lack of measured data. Additional measured neutron data is needed to bring MCNP simulations closer to detector measurements.

For some detector scenarios a best model will exist. CGMF’s slow run time prohibits it from regular use. FREYA’s angular correlation will improve results for low solid angle detectors, while PoliMi’s angular correlation was shown to worsen the agreement. Although FREYA improved measurement agreement with Cf-252, this is clearly because its Cf-252 neutron energy is 150 keV too high. The standard model has no angular correlation and its induced fission multiplicities are unphysical. There is no best model, so the standard model will likely be used to be consistent with historical results. The range of multiplicities between the models had a 1.8% effect on doubles rate, which was the smallest effect. The range of energies was up to 6% while in extreme cases the angular correlation effect was up to 30%. Hopefully these models will be quickly improved and these uncertainties can be eliminated.
CHAPTER 6
ENCAPSULATION EFFECTS ON THE NEUTRON SPECTRUM

Introduction

In this work the effects of neutron source encapsulation on the neutron spectrum were investigated. Measurements and simulations are often performed with Cf-252 neutron sources, which are widely available and well characterized. These sources are a convenient surrogate for other neutron emitters at nuclear facilities which may be unavailable to the companies or at the facilities where these detectors are constructed. A correction must be made to account for the change in energy between Cf-252 and the neutron source of interest [83, 84]. In this work the possibility of using source encapsulation as this correction was investigated. Instead of a calculation, placing the source in some material will modify the emitted neutron spectrum such that the detector response to the Cf-252 and encapsulation is identical to the response to a surrogate. Californium encapsulation was adjusted to produce a surrogate to a Pu-240 source in the Active Well Coincidence Counter [17]. Through this investigation, the related topic of approximations in neutron spectrum perturbation was also studied. Often in safeguards detectors and their simulations in MCNP, the encapsulation of neutron sources is often dismissed as ‘light’ and is not considered further. However there is no standard or guidance on what qualifies as a lightly encapsulated source. In this work the effects of encapsulation on neutron spectrum and intensity are quantified to resolve this dilemma and inform decisions on the meaning of light encapsulation. Common encapsulation

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materials and thicknesses, as well as commercial encapsulations, were studied in general and for a specific system.

The prompt fission neutron spectrum (PFNS) of a bare Cf-252 source is often approximated by analytical equations. For example, ISO 8529 recommends a Maxwell distribution with a temperature parameter of 1.42 MeV which generates a mean energy of 2.13 MeV [85]. A Watt spectrum is also used, with a temperature parameter of 1.175 MeV and the kinetic energy of fragments per nucleon of 0.359 MeV which generates a mean energy of $2.122\pm0.017$ MeV [80]. When the source encapsulation is dismissed as neutronically light, these representations are then the source spectrum.

A typical encapsulation is the Amersham X1, a stainless steel cylinder about 10mm long and a diameter of 7.8mm with a wall thickness of 1.6mm [86]. Presumably it would qualify as light, but its perturbation of the angular distribution from a near perfect isotropic is well established. The different thicknesses along the cylinder top and bottom create a cylindrically symmetrical angular distribution from the isotropic Cf-252 point source at the center [87]. This effect is not dismissed and instead is corrected for during calibrations of fluence detectors [88, 89]. Perhaps the impact on the neutron spectrum caused by neutron interactions in the source encapsulation is also relevant and should be accounted for. The detailed response function of the detector will also play a role. The degree of sensitivity to spectrum changes will control whether the difference between a 1mm and 3mm stainless steel container, or some other jacketing materials, matters or not.

**Calculations, Simulation and Measurements**

First, a simple analysis was performed to justify why encapsulation needs to be accounted for in neutron source measurements and to demonstrate that for common
source types the neutron spectrum is expected to vary linearly with wall thickness. The possible interaction pathways were considered to find an analytic solution to the emergent mean energy as a function of encapsulation wall thickness. Then, published values from the literature were used to confirm the calculations of this work. The values come from an attempt to deliberately moderate Cf-252 and AmBe (α,n) source spectra as an alternative to particle accelerator facilities for a calibration source of neutron dosimetry equipment. They show the variation of the mean energy as a function of spherical shell encapsulation.

An MCNP6.1.1b simulation of these results was used to show agreement between analytical theory, published work, and simulation [31]. This agreement gives confidence to additional modeling of the neutron gains, losses, average energy, and energy spectrum for spherical encapsulations of an array of potential materials. Commercial encapsulations were also simulated for this analysis. Most source encapsulation is in one of several standard forms from the manufacturer. They are often cylindrical in shape and made of 304L stainless steel, although Zircalloy-2 is sometimes used. There are also spacers and other differences in the inner source cavity void, and the use of threaded studs and other features. The default Cf-252 spontaneous fission energy spectrum was used, which is a Watt spectrum with \( a = 1.180 \) MeV and \( b = 1.03419 \) MeV\(^{-1}\). The mean energy was 2.13 MeV. Default physics options including non-analog transport were used because only total neutron counting was relevant. The neutrons were tallied over a sphere with a radius of 30cm, centered at the Cf-252 point source.
In real applications the full energy distribution is needed to understand the effect on a particular detector. The detection efficiency is the result of the energy distribution coupled to the detector response function. Cylindrical encapsulation was manufactured and measured in the AWCC, and used as verification of MCNP simulations. The cylinders are concentric shells that fit into each other like ‘Russian dolls’, Figure 6-1. Each has a 0.5 cm wall thickness and they were combined to make thicknesses of 0.5-2 cm. The smallest encapsulation was made to fit the Isotope Product Laboratories’ A7-869 Cf-252 source in an A3026 capsule. Copper, stainless steel, and polyethylene encapsulations were made. The emergent neutron spectrum was calculated for each thickness to show the effects of increasing thickness. Then the AWCC response function was simulated. The AWCC simulated with one polyethylene shell is shown in Figure 6-2. The AWCC was operated in fast mode with the cadmium liner and full plug rings. The liner is 1.6mm thick and absorbs neutrons below about 0.7 eV. The nickel reflector was used and the source was placed in the center of the cavity on a lab jack. The lab jack was raised or lowered to keep the Cf-252 material in the center of the detector as more encapsulation was added. The detector has two rings of He-3 tubes, and the ratio between the tubes acts as an indicator of the neutron energy. Simulations and measurements of the ring ratios were used to verify that the MCNP generated spectra of the encapsulations is accurate. The use of ring ratios eliminates dependence on the source strength, which has a relatively large uncertainty.
Finally, the encapsulation needed for Cf-252 to match the detection efficiency of Pu-240 and act as a surrogate was calculated. The relative efficiency of the manufactured encapsulations was also simulated and measured to increase confidence.
Results

As with any exponential expression with small change in the independent variable, the spectral indices of the emergent neutron spectrum are expected to vary linearly with encapsulation thickness at small thicknesses. For a point source encapsulated by a thin spherical shell it can be assumed that the only reaction of significance taking place is elastic scattering. If the encapsulation is sufficiently thin it can be assumed that only a single scatter per neutron will occur. The probability, $p_s$, that a neutron will scatter on its way out is given to first order by Equation 6-1, where $\Sigma_s$ is the macroscopic scattering cross section of the shell material and $t$ is the thickness.

$$p_s = \Sigma_s t \ll 1$$ (6-1)

Thus, the probability of neutrons emerging without scattering or suffering any energy loss is $1-p_s$. By the conservation of mass and momentum governing scattering kinematics, neutrons that scatter will lose half of the maximum energy that can be transferred to the target nucleus on average, assuming the scattering is isotropic in the center of mass reference frame. The mean fractional energy loss, $f$, is given by Equation 6-2, where $A$ is the ratio of mass of the target nucleus to a neutron. For an element $A$ can be approximated as the molar mass in g mol$^{-1}$.

$$f = \frac{2A}{(1+A)^2}$$ (6-2)

The average energy of the neutrons that scatter, $\bar{E}_s$, is then given by Equation 6-3 where $\bar{E}$ is the average energy of the emitting source.

$$\bar{E}_s = \bar{E}(1-f) = \bar{E}(1 - \frac{2A}{(1+A)^2})$$ (6-3)

The average emergent neutron energy of the encapsulated source $\bar{E}_e$ is the probable combination of the scattered and unscattered neutrons given by Equation 6-4,
which can be rearranged to give Equation 6-5. Given the assumptions of only single
scatters which is valid for a thin capsule, the emerging neutron energy is expected to
decrease linearly with capsule wall thickness. A real capsule has the potential for
additional interactions such as inelastic scatter will further decrease the emergent mean
energy. These interactions may also cause a gain or loss of neutrons through, for
example, \((n,2n)\) and \((n,\text{alpha})\) interactions respectively. The expected ratio \(R\) of the
emergent neutron energy to the initial energy before any interaction can then be given
by equation 6-6, for a thin encapsulation where \(b\) is a coefficient specific to the
composition and density of the wall material.

\[
\bar{E}_e = (1 - p_s)\bar{E} + p_s\bar{E}_s = (1 - p_s)\bar{E} + p_s\bar{E}(1 - \frac{2A}{(1+A)^2})
\]  
(6-4)

\[
\bar{E}_e = \bar{E}(1 - \frac{2A}{(1+A)^2} \Sigma_s t)
\]  
(6-5)

\[
R = \frac{\bar{E}_e}{\bar{E}} = 1 - bt
\]  
(6-6)

This result is compared to a study from Hsu and Chen where a Cf-252 point
source was encapsulated by spheres of a range of thicknesses and materials in an
try to generate spectra useful for the calibration of health physics detectors [90].
The studied materials were Be, graphite, Al, Fe, Cu, Pb, LiD, H2O, D2O, polyethylene,
glass, and concrete, at radii of 2.54, 5.08, 7.62, 10.16, 15.32, and 20.32 cm, which
correspond to whole numbers of inches. The resulting neutron spectra were computed
at 50cm from the center. However the results are of limited value. They are presented
graphically, making quantified evaluation difficult. Gains and losses are not analyzed
and the average energy is only given for one material, copper. The average energy with
0 copper thickness is given as 2.54 MeV, which is much greater than the 2.12 MeV
accepted value [80]. The error was suppressed in this work by normalizing the results to
the expected value. The data was fit to the general form in Equation 6-7, and at thin wall thicknesses fit the reduced form of Equation 6-6.

\[ R = e^{-bt} \]  

(6-7)

The parameter \( b \) was solved to be 0.079 cm\(^{-1}\) which resulted in an \( R^2 \) fit to the data of 0.99992, Figure 6-3. The strong agreement demonstrates the accuracy of the analytic argument. From the figure it can be seen that the limit to a ‘thin’ wall encapsulation is 1 to 2 cm, after which the change can no longer be approximated as linear. It can also be seen that a 21 keV or 1% change in average energy occurs at a wall thickness of 0.127mm of copper. This small amount of encapsulation produces a large change in a typical thermal neutron detector counting efficiency. The High Level Neutron Coincidence Counter gains about 17% detection efficiency per 1 MeV decrease in energy [22, 63]. The 21 keV lost to encapsulation results in a 0.36% increase in detection efficiency, which is significant relative to the typical measurement uncertainty for Cf-252 sources.

MCNP was used to simulate materials and thicknesses to calculate the relevant quantities, gains, losses, the net change in the emission of neutrons, and the average energy. Spectrum information was also collected for later use. The result for copper is shown in Figure 6-4, and the Hsu results are added and show good agreement. This gives confidence to the MCNP simulations. The MCNP given mean energy of Pu-240 spontaneous fission is 1.93 MeV. The copper results were used to calculate that a 1.39 cm copper wall thickness would change a Cf-252 source’s average energy to that of a bare Pu-240 source.
The results for all of the materials and thicknesses of 1 to 20 cm are shown in Figure 6-5. The gains, losses, and net values are per source neutron. Lead has the least effect on average energy which is to be expected due to its large Z. Likewise, polyethylene, with its large hydrogen content, has the greatest effect. Beryllium’s \((n,2n)\) reaction causes a 6.5% increase in the emitted neutrons at a thickness of 15cm, and also, interestingly, would turn a random neutron emitter like an \((\alpha,n)\) source into a coincidence neutron emitting source. Stainless steel has a negligible effect on net intensity but can significantly change the average energy at common encapsulation thicknesses.
Figure 6-4. Comparison of this work’s MCNP results for copper and that of Hsu and Chen. The results are normalized to remove the mean energy discrepancy of Hsu. Photo credit: Weinmann-Smith.

<table>
<thead>
<tr>
<th>Aluminum</th>
<th></th>
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</thead>
<tbody>
<tr>
<td>Spherical Radius (cm)</td>
<td>Gains</td>
<td>Losses</td>
<td>Net</td>
<td>Average energy (MeV)</td>
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<td>Gains</td>
<td>Losses</td>
<td>Net</td>
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Figure 6-5. Continued
### Figure 6-5. Continued

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<td>2.91*10^-3</td>
<td>9.97*10^-1</td>
</tr>
<tr>
<td>3</td>
<td>8.88*10^-5</td>
<td>4.34*10^-3</td>
<td>9.96*10^-1</td>
</tr>
<tr>
<td>4</td>
<td>1.12*10^-4</td>
<td>5.77*10^-3</td>
<td>9.94*10^-1</td>
</tr>
<tr>
<td>5</td>
<td>1.32*10^-4</td>
<td>7.18*10^-3</td>
<td>9.93*10^-1</td>
</tr>
<tr>
<td>10</td>
<td>2.06*10^-4</td>
<td>1.45*10^-2</td>
<td>9.86*10^-1</td>
</tr>
<tr>
<td>15</td>
<td>2.43*10^-4</td>
<td>2.39*10^-2</td>
<td>9.76*10^-1</td>
</tr>
<tr>
<td>20</td>
<td>2.64*10^-4</td>
<td>3.87*10^-2</td>
<td>9.61*10^-1</td>
</tr>
</tbody>
</table>

Figure 6-5. Neutron effects from encapsulation of various materials and thicknesses. Photo credit: Weinmann-Smith.

While the results of spherical encapsulation are interesting, the calculations of standard manufacturer’s encapsulations are more practical. The A3026 capsule is provided by Eckert & Ziegler [91]. The FTC capsules come from Frontier Technology Corporation, where s denotes a shorter version of the capsule [92]. FTC 10 is singly encapsulated while FTC 100 includes a second encapsulation around FTC 10. FTC 10
and FTC 100 are identical to Savannah River National Laboratory’s SR-Cf-1X and SR-CF-100 capsules respectively. Amersham, now known as QSA, makes the X1 capsule [93]. Physical characteristics of the capsules are given in Table 6-1, while Table 6-2 shows the gains, losses, net neutrons, and average spectrum energy. The difference between 304 and 304L stainless steel is the carbon content, a less than 1% overall change. It is therefore not surprising that the difference between the two was found to be negligible. Any absorption and gains are also negligible, but the range of emerging neutron energies is 46 keV, and the A3026 capsule causes a 53 keV decrease from a bare Cf-252 source, which has been shown in this work to be significant and measureable in common detectors.

Table 6-1. Common manufacturer encapsulation’s physical characteristics

<table>
<thead>
<tr>
<th>Capsule</th>
<th>Material</th>
<th>Mass (g)</th>
<th>Outer diameter (cm)</th>
<th>Outer length(cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A3026</td>
<td>304 SS</td>
<td>18.4</td>
<td>0.942</td>
<td>3.60</td>
</tr>
<tr>
<td>FTC 10s</td>
<td>304L SS</td>
<td>1.7</td>
<td>0.551</td>
<td>1.19</td>
</tr>
<tr>
<td>FTC 10</td>
<td>304L SS</td>
<td>2.9</td>
<td>0.551</td>
<td>2.46</td>
</tr>
<tr>
<td>FTC 100</td>
<td>304L SS</td>
<td>15.9</td>
<td>0.942</td>
<td>3.76</td>
</tr>
<tr>
<td>Amersham X1</td>
<td>SS</td>
<td>3.1</td>
<td>0.782</td>
<td>0.98</td>
</tr>
</tbody>
</table>

Table 6-2. Common manufacturer encapsulation’s effects on the emergent neutrons

<table>
<thead>
<tr>
<th>Capsule</th>
<th>Gains</th>
<th>Losses</th>
<th>Net</th>
<th>Average energy (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A3026</td>
<td>1.816*10^-5</td>
<td>7.563*10^-4</td>
<td>9.993*10^-1</td>
<td>2.067</td>
</tr>
<tr>
<td>FTC 10s</td>
<td>5.643*10^-6</td>
<td>2.221*10^-4</td>
<td>9.998*10^-1</td>
<td>2.112</td>
</tr>
<tr>
<td>FTC 10</td>
<td>5.111*10^-6</td>
<td>2.072*10^-4</td>
<td>9.998*10^-1</td>
<td>2.113</td>
</tr>
<tr>
<td>FTC 100</td>
<td>1.411*10^-5</td>
<td>5.760*10^-4</td>
<td>9.994*10^-1</td>
<td>2.083</td>
</tr>
<tr>
<td>Amersham X1</td>
<td>1.012*10^-5</td>
<td>4.476*10^-4</td>
<td>9.996*10^-1</td>
<td>2.100</td>
</tr>
</tbody>
</table>

So far only the average energy has been shown as a result. Since a neutron detector response function is complex and nonlinear, knowing the difference in average energy is not sufficient to solve for the change in detection efficiency. For example, neutrons half at 0.5 MeV and half at 1.5 MeV will have a different efficiency than
neutrons all at 1 MeV for a detector with a peak efficiency at 1 MeV. Recall that a Cf-252 point source placed in a copper sphere with radius of 1.39 cm will match the average energy of Pu-240. The energy spectrum of all three of these sources is shown in Figure 6-6. The difference in the shapes of the spectra can be clearly seen. This demonstrates that to use encapsulation to create a surrogate source, more detailed analysis is necessary. Such an analysis is detector specific because it depends on the individual detector response function. To validate the analysis measurements of Cf-252 in different encapsulations were taken. The simulated results of the emerging neutron spectrum from the encapsulation are shown in Figs. 6-7 through 6-10. The first figure’s scale is log-log. Increasing encapsulation thickness increases the change in spectrum for all materials. The polyethylene spectrum clearly stands out from copper and stainless steel. The buildup at near thermal energies signals a relatively bimodal distribution caused by the population of neutrons that scatter on hydrogen and lose potentially all of their energy, and on average half. The absorption resonances in copper at 2 keV and other energies are also clear.

The AWCC response function, which is the efficiency as a function of energy, is shown in Figure 6-11 and is calculated through simulation. The complexity clearly demonstrates why average neutron energy is insufficient for calculate detection efficiency. The AWCC has two rings of He-3 tubes, Figure 6-12, at different polyethylene depths. Each has an independent response function and they reduce the detector dependency on neutron energy, as higher energy neutrons that pass the inner ring are more likely to be absorbed by the outer ring. Single ringed neutron detectors lack this buffering effect while more than 2 rings amplify it. The response function shows
a peak efficiency at 1.2 MeV neutron energy for the detector as a whole while the inner ring peak efficiency is at 0.6 MeV and the outer ring peak efficiency is at 2.1 MeV. The measured and simulated ring ratios are shown in Figure 6-13.

Figure 6-6. Neutron spectra for Cf-252, Cf-252 in copper, and Pu-240. Photo credit: Weinmann-Smith.
Figure 6-7. Exit spectra of encapsulation. Photo credit: Weinmann-Smith.

Figure 6-8. Exit spectra of polyethylene. Photo credit: Weinmann-Smith.
Figure 6-9. Exit spectra of stainless steel. Photo credit: Weinmann-Smith.

Figure 6-10. Exit spectra of copper. Photo credit: Weinmann-Smith.
Figure 6-11. AWCC response function for the inner and outer rings, and the detector as a whole. Photo credit: Weinmann-Smith.

Figure 6-12. Top down MCNP simulation of the AWCC. Photo credit: Weinmann-Smith.
The results were normalized to the ratio of the A3026 encapsulated source without additional encapsulation to remove bias in the simulation of the detector. The relative statistical uncertainty was less than 0.02% for measurements and 0.09% for simulations, and is too small to be seen in the figure. They agree within 2 standard deviations for all cases but 2cm of copper, which indicates that the simulations are accurate with regards to the emitted neutron energy spectrum and the AWCC model.

Finally, changes in detection efficiency by the AWCC of various encapsulations were measured and simulated. The encapsulation necessary for a Cf-252 source to match the Pu-240 source was also simulated. Again measurements were used to validate simulation results so that the simulations could be used when measurements are impossible, like measuring bare Pu-240 or bare Cf-252 for example. Again the results are normalized to the A7-869 source in the A3026 capsule to account for the
unknown source strength. The results are shown in Figure 6-14 with statistical uncertainties of 0.07% for simulations and 0.02% for measurements, which are both too small to plot. Note that the y-axis range is only 16% of the efficiency of the A3026 encapsulated source.

First, the close agreement between measurements and simulations of poly, copper, and steel encapsulations gives confidence to the results. The only significant divergence is polyethylene at large thicknesses, and the large uncertainties in polyethylene cross sections are discussed elsewhere in this work. Starting from a bare Cf-252 point source, the detection efficiency is simulated to be 0.98 that of the A3026 encapsulated source. The difference between the absolute efficiency of bare Cf-252 and bare Pu-240 is 0.23%. Adding the A3026 encapsulation increases the efficiency by about 2%. However the Pu-240 relative detection efficiency is 0.992 and so the A3026 encapsulation overshoots the target efficiency. The detector is under moderated at this energy spectrum and so adding additional moderation in the form of copper or steel will only increase the detection efficiency further, up to about 2%. Additional polyethylene over moderates the detector and is demonstrated to decrease the efficiency. A 0.5cm shell thickness around the A3026 capsule decreases the relative efficiency to 0.986, and the absolute detection efficiency of this configuration is 0.13% lower than bare Pu-240. To match the Pu-240 detection efficiency a polyethylene shell of 0.35cm around the A3026 capsule is needed. The relative efficiencies are both 0.992 and the absolute efficiencies differ by only 0.004%, which is within one standard deviation of the statistical uncertainty. Thus with respect to detection efficiency a Cf-252 source in an
A3026 capsule and 0.35cm polyethylene wall thickness of additional moderation will match a Pu-240 source in the AWCC exactly.

![Graph](image)

Figure 6-14. AWCC efficiencies relative to the A7-869 source in the A3026 capsule. Photo credit: Weinmann-Smith.

**Conclusions**

Radiation sources are almost always encapsulated in some material to prevent the spread of contamination. In the neutron measurement and assay of these sources the encapsulation is often deemed ‘neutronically light’ and its effects ignored. This work has demonstrated that even a few millimeters of metallic encapsulation perturbs the emitted neutron spectrum to a readily measureable degree. The effects of common encapsulations and many materials that may perturb emitted neutrons were shown to guide users to decide what can be considered neutronically light and what must be accounted for. Encapsulation perturbations can also be taken advantage of to lower a sources average energy, adjust its spectrum, and to match the detection efficiencies of
other sources. This was done in the Active Well Coincidence Counter, a common safeguards system with a nonlinear neutron response. An additional 0.35 cm of polyethylene around an A3026 encapsulated Cf-252 source was shown to match the detection efficiency of a bare Pu-240 spontaneous fission source. This encapsulation would allow Cf-252 to be used as a widely available surrogate in the testing and calibration of detectors when most restricted Pu-240 sources are not available.
CHAPTER 7
SUMMARY AND FUTURE WORK

Neutron multiplicity counting is a well-established technique used to assay the fissile mass of special nuclear material for international safeguards. Application of the technique is very much ‘engineering’ in that the systems work well, but calibration is required and knowledge of the underlying data is lacking such that scientific calculations to solve the systems are not accurate. The implementation of these detectors was adequate for the requirements, accuracies, and counting times, for the IAEA to verify compliance with the NPT. However, every year the material and facilities under safeguards continues to grow and so does the burden on IAEA inspectors. Improvements to techniques with better accuracy and counting times, and reduced burden on design, calibration, implementation, and operation teams are needed to maintain capabilities and free resources for other applications. Simulation accuracy is the most accessible target for these improvements. In the past, simulations were used to show general trends and approximate effects. Accuracies of 10% were sufficient and accepted. As time has continued and hands on nuclear work has become more expensive the reliance on simulation has increased. As technology has advanced new capabilities measure with better accuracy and computing power has become inexpensive. Computing and simulation has revolutionized other fields. In the next generation of safeguards tools accuracies of 1% or better should be expected, and are attainable.

This work addressed some of the limiting uncertainties in detection and simulation accuracy. The result is a step closer towards high accuracy simulations.
Some of the results were specific to one application while others apply to all types of multiplicity counting.

The accuracy of cosmic ray simulations was quantified and the simulations generally agreed with the measured results. Cosmic rays limit the uncertainty that can be reached in measurements of low activity samples, as the cosmic ray signal hides the radiation source signal. The simulations were used to identify cosmic ray characteristics that are impossible to measure. These characteristics revealed trends that may be useful in future cosmic ray rejection techniques. The cosmic ray induced multiplicity was shown to greatly exceed that of non-multiplying fission sources. This isolated measurement of only cosmic rays is correlated to cosmic rays at lower multiplicities, which are a combination of cosmic and source neutrons. Algorithms and methods to leverage this phenomenon are currently under development by others. Ultimately the information uncovered in this work may lead to improved accuracy in some neutron counting applications.

In the simulation of active interrogation measurements the AmLi spectrum is often assumed to be the author’s preferred published in the literature. For the first time, 17 AmLi sources were measured and the variations in neutron spectra between them were quantified. This result is valuable in uncertainty quantification and sensitivity analysis, which is used, among other things, to prioritize improvements in data. Additionally, spectra were generated to accurately reproduce the measurements. The spectra were generated for manufacturer-specific averages of multiple sources, which provides a new resource which will reduce uncertainty for researchers as they can choose a spectrum more likely to match their source. Additionally, the collected data
allow LANL to develop a spectrum for any of their sources, eliminating one source of uncertainty.

The fission neutron data used in simulations is one of the largest sources of uncertainty. The new physics based fission models being released in MCNP6.2 have the potential to greatly reduce this uncertainty. These fission models were compared to measurements of nuclear data and of complex safeguards detection systems to quantify improvement with the goal of identifying the best model for future use in the safeguards community. None of the fission models clearly reproduced the measurements in all cases. While a best model was not found, instead the work identified exactly what nuclear data the models failed to reproduce. Areas where nuclear data was lacking were also identified. The impacts of differences in the models on safeguards detectors were found. For example, the neutron angular correlation had the largest effect. Measurements to clarify nuclear data uncertainties are needed, and incorporating the results in future MCNP simulations will increase their accuracy.

In simulations of neutron detectors detailed physical descriptions are needed, but the level of detail is not well known. The effects of thin source encapsulations were studied to clarify the effects of a few millimeters of metal between the source and detector. Many materials and thicknesses were investigated to provide users with an estimate for their configuration. The results again apply to uncertainty quantification and will help users to identify the level of detail needed in their simulations, and the effects they can expect in measurements. The method for developing surrogate sources which have the same detection efficiencies as another source by adjusting their energy spectrum through encapsulation was also demonstrated. The polyethylene thickness
required for a Cf-252 source to act as a Pu-240 source for total neutron counting was found. This enables users to make measurements when Pu-240 sources are not available.

While 1% accuracy will require a broad effort across many organizations, some work remains in the small scale domain. Full 1% accuracy for all of neutron multiplicity counting requires several facets of nuclear data for as many as 15 neutron sources. However, a well characterized simple detection scenario with minimal aspects would narrow the scope of necessary data. Efforts could be focused on this configuration, and when it is fully solved, small changes could be used to expand the scope to additional sources, materials, and applications. Such a scientific approach is regularly used in the criticality safety community, and should be applied to safeguards.

Although it is simple, accurate measurements of polyethylene density for existing detectors will have the largest improvement in simulation accuracy. There is a large range of published values and manufacturer statements should be verified for a specific detector. In the near future these measurements will be carried out with a target accuracy of 0.1% uncertainty to better inform simulation. Then the physical model of the detector should be refined to great detail. This should include any source encapsulations, detector electronics, the detector frame, He-3 tube structure and any other small details that are often overlooked. Reducing uncertainties introduced by the physical description of the detector will make the study of nuclear data more impactful and effective.

The progress described in this work, and successful execution of the future work, will result in simulations that accurately predict measurement results absolutely. This
will enable great increases in efficiency, as new detector designs and applications are conceptualized and tested in days instead of years. Detailed calibrations will no longer cost millions of dollars and require the manufacture, characterization, and measurement of nuclear material. This new capability in combination with the improvements to measurements discussed with respect to cosmic rays and encapsulation will result in a more agile and capable IAEA, increasing confidence in the compliance of nations around the world with the Nonproliferation Treaty.
APPENDIX
LIST OF PUBLICATIONS

The following publications are a result of the four projects of this work.


The following publications are a result of other related projects completed over the course of this work.


LIST OF REFERENCES


[85] International Organization for Standardization, Reference neutron radiations -- Part 1: Characteristics and Methods of Production, ISO 8529-1:2001-02-01(E), see also ISO 8529-1:2001/Cor 1:2008(F) which provide a typographical correction to Table A.4 the \(^{241}\text{Am-Be(}\alpha,\text{n)}\) spectrum, 2001.


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BIOGRAPHICAL SKETCH

Robert Weinmann-Smith was raised in west Florida. He was first introduced to nuclear radiation through a high school course. He joined the nuclear engineering department at the University of Florida and graduated with a bachelor’s degree in 2014, with research in nuclear materials. Through a 2014 summer internship at Los Alamos National Laboratory he became interested in radiation detection. He continued to work with LANL through graduate school, moving to Los Alamos year round in 2016. Robert earned his Ph.D. in 2018 from the University of Florida.