Evaluation of Am-Li Neutron Spectra Data for Active Well Type Neutron Multiplicity Measurements of Uranium

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Abstract

Safeguarding nuclear material is an important and challenging task for the international community. One particular safeguards technique commonly used for uranium assay is active neutron correlation counting. This technique involves irradiating unused uranium with (α, n) neutrons from an Am-Li source and recording the resultant neutron pulse signal which includes induced fission neutrons. Although this non-destructive technique is widely employed in safeguards applications, the neutron energy spectra from an Am-Li sources is not well known. Several measurements over the past few decades have been made to characterize this spectrum; however, little work has been done comparing the measured and theoretical spectra of various Am-Li sources to each other. This paper examines fourteen different Am-Li spectra, focusing on how these spectra affect simulated neutron multiplicity results using the code Monte Carlo N-Particle eXtended (MCNPX). Two measurement and simulation campaigns were completed using Active Well Coincidence Counter (AWCC) detectors and uranium standards of varying enrichment. The results of this work indicate that for standard AWCC measurements, the fourteen Am-Li spectra produce similar doubles and triples count rates. The singles count rates varied by as much as 20% between the different spectra, although they are usually not used in quantitative analysis, being dominated by scattering which is highly dependent on item placement.

Introduction

Nuclear safeguards methods are used by the international community to ensure that nuclear materials are not used for illicit purposes. These methods consist of techniques such as item accounting, bulk material accounting, containment and surveillance, environmental sampling, etc. [1]. One particular material accounting technique used for uranium materials is active neutron correlation counting. This

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technique consists of irradiating fresh uranium with (α, n) neutrons from an Am-Li source and measuring the time-correlated fission neutrons from the uranium. Most commonly, coincidence (i.e., doubles) counting is applied, but higher order multiplicity (i.e., triples) counting has also been used [2]. Am-Li sources are ideally suited for this technique because they do not emit neutrons in time-coincidence (i.e. they are only emitted as single neutron events) and have neutron energies concentrated below the approximate 1 MeV fission threshold for ^{238}U [3]. This allows the measurement technique to largely focus on ^{235}U fissions amplified by multiplication in the whole uranium sample.

The neutron energy spectrum from an Am-Li source is continuous, stretching from 0 to approximately 1.5 MeV, with a mean energy of approximately 0.5 MeV. ^{241}Am has an average alpha energy of 5.48 MeV, which is above the ^7Li (92.5% abundance) (α, n) threshold energy of 4.38 MeV. This is not true for ^6Li (7.5% abundance), which has an (α, n) threshold energy of 6.32 MeV, thus preventing it from contributing to the Am-Li neutron energy spectrum.

The chemical and physical composition of Am-Li sources is often a finely mixed powder of AmO_2 and LiH . Because there are oxygen atoms in close proximity to ^{241}Am , the Am-O (α, n) neutrons also contribute to the Am-Li neutron energy spectra. The Am-O neutron energy spectrum is also continuous, but stretches from 0 to approximately 4.6 MeV, with a mean neutron energy of approximately 2 MeV. The yield of Am-O neutrons is several orders of magnitude smaller than that of Am-Li, depending on the ratio of AmO_2 and LiH powders [4].

Impurities with low atomic mass, such as beryllium and fluorine, can also affect the neutron energy spectra from an Am-Li source; however, the purity of the AmO_2 and LiH powders is usually such that this contribution can be ignored. The last significant factor which can affect the Am-Li neutron energy spectrum is moderation due to hydrogen and oxygen atoms in the sample itself. Neutron moderation with these elements can reduce the neutron energies before they exit the source container [4], resulting in a softened energy spectrum.

When developing new techniques, equipment, or extending calibrations, it is often preferable and advantageous to employ extensive computer simulations to reduce the cost of performing burdensome physical measurements. A key component to high-fidelity simulations is accurate nuclear data, including radiation source terms [5]. The Am-Li source term is the topic of this research. Several measurements of the neutron energy spectrum from an Am-Li source have been made over the past few decades. The thirteen most notable measurements and calculations are shown in Table 1. Normalized Am-Li neutron energy spectra from these thirteen sources are shown in Figs. 1-13.

Table 1. Summary of Am-Li source data used in the MCNPX simulations.

Am-Li spectrum	Neutron source	Mean neutron energy [MeV]	Highest neutron energy [MeV]	Number of data points	Features or comment
H. Werle (1970) [6]	Am-Li	0.433	1.6	160	Hydrogen proportional counter
R. Birch et. al.	Am-Li	0.445	1.5	30	Hydrogen filled proton

(1984) [7]					recoil counter
L. Trykov et. al. (1997) – 2 mm W [8]	Am-Li + Am-O	0.459	4.0	21	Bonner sphere 0.01 – 4 MeV Proton recoil 0.4 – 4 MeV
H. Delafield and R. Birch (1989) [9]	Am-Li	0.474	1.6	27	
National Physical Laboratory [10]	Am-Li	0.478	2.0	236	Bonner sphere measurement methodology
H. Ing et. al. (1981) [11]	Pu-Li	0.485	4.0	42	Hydrogen filled proton recoil counter
P. Rinard (2011) [12, 13]	Am-Li + Am-O	0.492	3.5	38	Bonner sphere measurement methodology
L. Trykov et. al. (1997) – 4 mm W [8]	Am-Li + Am-O	0.519	4.0	21	Bonner sphere 0.01 – 4 MeV Proton recoil 0.4 – 4 MeV
J. Owen et. al. (1981) [14]	Am-Li	0.532	1.665	74	Pb shielded high pressure ³ He spectrometry
K. Geiger and L. Van der Zwan (1971) [15]	⁷ Li(α ,n) ¹⁰ B	0.536	1.52	38	Threshold (5.51 MeV) below 1 st Excited state
H. Tagziria and M. Looman (2012) [16]	Am-Li	0.558	2.0	50	Variety of detectors with unfolding techniques
D. Weaver et. al. (1982) [17]	Am-Li	0.559	1.98	88	³ He spectrometry
SOURCES4A (1999) [18]	Am-Li + Am-O	0.583	6.0	24	GNASH statistical model calculation

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72 Figure 1. Normalized Am-Li neutron energy spectra from H. Werle (1970).

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Figure 2. Normalized Am-Li neutron energy spectra from R. Birch et. al. (1984).

Figure 3. Normalized Am-Li neutron energy spectra from L. Trykov et. al. (1997) – 2 mm W.

Figure 4. Normalized Am-Li neutron energy spectra from H. Delafield and R. Birch (1989).

Figure 5. Normalized Am-Li neutron energy spectra from National Physical Laboratory.

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Figure 6. Normalized Am-Li neutron energy spectra from H. Ing et. al. (1981).

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Figure 7. Normalized Am-Li neutron energy spectra from P. Rinard (2011).

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Figure 8. Normalized Am-Li neutron energy spectra from L. Trykov et. al. (1997) – 4 mm W

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Figure 9. Normalized Am-Li neutron energy spectra from J. Owen et. al. (1981).

Figure 10. Normalized Am-Li neutron energy spectra from K. Geiger and L. Van der Zwan (1971).

Figure 11. Normalized Am-Li neutron energy spectra from H. Tagziria and M. Looman (2012).

Figure 12. Normalized Am-Li neutron energy spectra from D. Weaver et. al. (1982).

Figure 13. Normalized Am-Li neutron energy spectra from SOURCES4A (1999).

These Am-Li spectra have a similar general shape, but their exact values can vary significantly. Based on a review of the literature, computer simulations in which an Am-Li neutron energy spectrum is required often only use one of the above spectra in the simulation [19]. When simulations are performed with multiple different Am-Li spectra, it is common to use only a few of the above spectra [13, 16]. No published simulations have been performed using all thirteen of the above spectra. Due to the lack of a comprehensive and systematic study in the literature and to support accurate radiation source term for high-fidelity measurements, experiments and computer simulations were performed using all candidate spectra. The goal of these measurements and simulations were to quantify the effect of these different spectra on the results of active neutron coincidence and multiplicity counting measurements on uranium materials.

Experiment

Two measurement campaigns were completed using active well type neutron coincidence counters measuring various enriched uranium standards. These campaigns were carried out at the PERFORMANCE Laboratory for Safeguards (PERLA) at the Joint Research Center (JRC) in Ispra, Italy in December 2010 and at the Safeguards Laboratory at Oak Ridge National Laboratory (ORNL) in Oak Ridge, Tennessee, in the summer of 2008. The purpose of the experiments was to provide measured data against which the predictions could be compared both in an absolute and a relative sense. However, the absolute

comparisons are of limited value because the Am-Li source strengths are relatively poorly known in contrast to the precision needed. On the other hand, the relative behaviours are valuable in demonstrating and justifying the use of Monte Carlo Simulations in extending calibrations to other materials and geometries.

JRC

The neutron detector used to perform the measurements at the JRC was the Canberra model JCC-51 Active Well Coincidence Counter (AWCC). This detector can be operated in an active configuration in which two Am-Li sources located in each end plug interrogate a uranium standard located in the cylindrical measurement cavity. The response of this detector is created by $^3\text{He}(n,p)^3\text{H}$ reactions in 42, 1 inch diameter, 4 atm ^3He tubes arranged in two concentric rings embedded in high density polyethylene (HDPE). The AWCC has a neutron die-away time are approximately 50 μs and a detection efficiency of approximately 33% for neutrons created in the measurement cavity with fission spectrum energies [20]. Four high enriched uranium (HEU) standards, three cans of U_3O_8 and one can of uranium metal, were measured. For all measurements the HEU standards were placed in the center of the measurement cavity using a thin aluminium stand. The mass and enrichment of each uranium standard is given in Table 2 [21].

Table 2. Composition, mass, and enrichment of uranium standards measured at the JRC.

Item name	Material composition	U mass [g]	^{235}U [wt.%]
Metal HEU	Metallic	140.5	36.1
90% HEU	U_3O_8 powder	46.39	89.90
36% HEU	U_3O_8 powder	169.39	36.20
20% HEU	U_3O_8 powder	169.45	20.00

Properties of the Am-Li sources used were estimated from documentation. Each source consists of approximately 0.4 g of Am_2O_3 powder mixed with 1.6 g of LiH powder for a combined powder density of 0.54 g/cm^3 . The total neutron emission rate from the combined sources is estimated to be 113,900 n/s $\pm$ 1.5%. This estimate was determined using cross-calibration with a similar National Physical Laboratory certified source. Each uranium standard was individually measured in the center of the measurement cavity of the AWCC in a “fast mode” configuration in which a cadmium liner was placed in the measurement cavity but with the nickel reflector removed. The “thermal mode” configuration was not used due to requirements of other research projects utilizing data from these measurements. The HDPE rings in the top and bottom end plugs were also removed. Measurement times consisted of 1 hour (60 cycles at 60 seconds per cycle) for each uranium standard, except for the 20% HEU standard, which was measured for 1.5 hours (90 cycles at 60 seconds per cycle). A background measurement (with the Am-Li sources still in the end plugs) was made for 5.6 hours (100 cycles at 200 seconds per cycle). The detector’s response for all measurements was recorded using a JSR-14 shift register [22] and the International Atomic Energy Agency Neutron Coincidence Counting (INCC) software program version

5.04, with the default quality control checks turned on [23, 24]. Pre-delay and gate width values for all measurements were set to 4.5 μ s and 64 μ s, respectively, which are the standard settings used by international inspectors.

ORNL

The neutron detector used to perform the measurements at ORNL was a Large Volume Active Well Coincidence Counter (LV-AWCC) design in which the diameter of the central cavity was expanded from 23 cm to 28 cm to accommodate larger diameter samples. In order to maintain approximately the same detector efficiency, the number of 1-inch diameter, 4 atm ^3He tubes was also increased from 42 tubes to 48 tubes. The neutron detection efficiency and die-away time are approximately 34%, depending on configuration, and 50 μ s respectively [21, 25].

Properties of the Am-Li sources used in the LV-AWCC were estimated from geometry and source certificate documentation. Each source consists of approximately 0.4 g of AmO_2 (1.2 Ci $\pm$ 1% of ^{241}Am) powder mixed with 1.6 g of LiH powder for a combined powder density of 0.5 g/cm³. The neutron emission rate of the sources was estimated from measurements of the Am-Li sources to be 48,850 and 49,950 n/s $\pm$ 3.0%. The measured uranium standards included three cans of U_3O_8 powder, each with approximately 194 g of uranium, at ^{235}U wt.% enrichments of 20.11%, 52.49%, and 93.17% [26]. Each uranium standard was individually measured in the center of the measurement cavity for the LV-AWCC in an active configuration for two non-consecutive 1-hour measurements (360 cycles at 10 second per cycle). The INCC program version 4.05, with default quality control checks turned on was used. A background measurement was performed for 1 hour (360 cycles at 10 second per cycle) with the detector in an active configuration. Pre-delay and gate width values for all measurements were set to 4.5 μ s and 64 μ s, respectively, which are the established values for routine use.

MCNPX Simulation

Detector design and development rely heavily on high-fidelity computer simulation to reduce the cost of performing many physical measurements. However, the accuracy of computer simulations is limited by the nuclear data fed into them. The neutron energy spectrum from an Am-Li source is one such data source. In order to understand how different measured Am-Li spectra affect active neutron multiplicity measurements of uranium using well type detectors, Monte Carlo N-Particle eXtended (MCNPX) version 2.7 simulations were performed. MCNPX is a nondeterministic code which uses Monte Carlo methods to determine the radiation transport for such particles as neutrons through a 3-dimensional combinatorial geometry and material composition. During the process of transporting radiation, MCNPX can tally various parameters of interest, including the time dependent $^3\text{He}(n,p)^3\text{H}$ reaction rate in the ^3He tubes, which can be used to calculate the correlated count rates in a detector [27-29].

Fourteen MCNPX models were created for each of the measurements performed at the JRC and ORNL. The geometry for these models included the detector, uranium standard, Am-Li sources with tungsten containers, concrete floor under the detector, and dry air inside the detector. The neutron source term consisted of the above mentioned thirteen neutron energy spectra and a fourteenth spectrum consisting of mono-energetic neutrons at 500 keV. The mono-energetic 500 keV source was used in the simulations because it represents perhaps the simplest approximation of neutron energies from an Am-Li source. The cross-section libraries for all of the models were from the Evaluated Nuclear Data Files (ENDF) 7 [30]. All fourteen models were run for 8×10^7 histories (Am-Li neutrons). The results tallied from these simulations were the singles, doubles, and triples count rates resulting from the time correlated $^3\text{He}(n,p)^3\text{H}$ reaction rate in the ^3He proportional counters. During the modelling process, several assumptions were made about the sources of neutrons. It was assumed that neutrons from cosmic-ray interactions [31], spontaneous fission in both the uranium and Am-Li materials [32], the U-O (α,n) reactions [32], and low atomic mass impurities in the Am-Li sources are all negligible. It was also assumed that for the measurement setup, photons would have a negligible effect on the detector's response. This has been determined in prior work and is achieved by suppression of the 60 keV gamma-rays of ^{241}Am by shielding and the high γ/n discrimination of the ^3He proportional counters [20].

In order to better understand how Am-Li neutrons of different energies contribute to the different count rates, another series of MCNPX simulations were performed with uniform probability distributions for different neutron energy groups. The energy bins of these 25 energy groups can be seen in Table 3. The detector and uranium standard chosen for these simulations was the JRC AWCC with the 90% HEU standard. Each simulation (energy group) was also run for 8×10^7 histories using a pre-delay and gate width of 4.5 μs and 64 μs , respectively.

Table 3. Energy bins used in the MCNPX energy group simulations.

0 – 0.01 eV	0.1 – 1 keV	40 – 50 keV	90 – 100 keV	500 – 600 keV
0.01 – 0.1 eV	1 – 10 keV	50 – 60 keV	100 – 200 keV	600 – 700 keV
0.1 – 1 eV	10 – 20 keV	60 – 70 keV	200 – 300 keV	700 – 800 keV
1 – 10 eV	20 – 30 keV	70 – 80 keV	300 – 400 keV	800 – 900 keV
10 – 100 eV	30 – 40 keV	80 – 90 keV	400 – 500 keV	900 – 1000 keV

Results

The results of the measured and simulated singles, doubles, and triples count rates can be compared in two main ways. First, all the simulated values can be compared to the measured results. This type of comparison is useful for an absolute comparison to the MCNPX simulations, but is less informative if the measured data has large uncertainties, e.g., the emission rate of the Am-Li sources is uncertain. In particular, in combination with other modelling uncertainties (acceptable differences between design and fabrication) it is not possible to pick a best spectrum based on agreement with the measurement. The second way to compare the results is to see how the simulation results compare to each other. This

type of comparison is useful to quantify relative changes in the MCNPX simulations and can determine which Am-Li spectra generate results which are outliers; this does not necessarily imply that the outlier is wrong. The spread in the results is informative about how well we know and describe the Am-Li spectrum in simulation space. The measured and simulated singles, doubles, and triples count rates are shown in Figs. 14-16 for the JRC results and Figs. 17-19 for the ORNL results. The general results reveal that the singles count rates are statistically different for the measurements and simulations; while the doubles and triples count rates are mostly within two standard deviations of each other. The data show a small, but non-zero, doubles count rate for the empty cavity measurements. This is due to induced fissions in the ^{241}Am present in the Am-Li sources, which is usually negligible for item counting in routine measurement cases.

JRC

The measured doubles and triples count rates from the JRC data match the average of the simulations within two standard deviations, with the exception of the 36% HEU doubles count rate. Although this implies the measured and simulated results are statistically the same for the doubles and triples count rates, it is difficult to draw conclusions due to the large statistical uncertainties in the measured data. The simulated doubles and triples count rates have significantly smaller uncertainties than their measured counterparts. This is a common challenge for practical applications of the AWCC for low mass samples, where the correlation signal is small and must be separated from accidental coincidences. Most of the simulated values are within two standard deviations of the simulation average. Expectedly, the singles count rates show far less agreement with values ranging from 13,000 to 17,000 c/s.

Fig. 14. Measured and simulated singles count rates from the JRC AWCC, without uncertainties.

Fig. 15. Measured and simulated doubles count rates from the JRC AWCC, with 1 sigma uncertainties.
 $\pm 1.5\%$ uncertainty in the Am-Li neutron sources strength not shown.

Fig. 16. Measured and simulated triples count rates from the JRC AWCC, with 1 sigma uncertainties. $\pm 1.5\%$ uncertainty in the Am-Li neutron sources strength not shown. The error bars on the measured data have been removed because they are too large to clearly read in the figure.

ORNL

The results from the ORNL data are qualitatively similar to those taken at the JRC. The main difference in the results is that the uncertainties in the measured doubles and triples count rates are smaller due to larger mass uranium standards, longer counting times, and few singles counts directly from the Am-Li sources. This provides more confidence that the simulations are precisely modelling the measurements.

268 As with the JRC results, the singles count rates vary considerably among the measurements and
 269 simulations, ranging from 5,500 to 7,500 c/s.

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 272 Fig. 17. Measured and simulated singles count rates from the ORNL LV-AWCC, without uncertainties.

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Fig. 18. Measured and simulated doubles count rates from the ORNL LV-AWCC, with 1 sigma uncertainties. $\pm 3\%$ uncertainty in the Am-Li neutron sources strength not shown.

Fig. 19. Measured and simulated triples count rates from the ORNL LV-AWCC, with 1 sigma uncertainties. $\pm 3\%$ uncertainty in the Am-Li neutron sources strength not shown.

Spectrum mean energy

In order to better understand how the mean energy of the neutron spectra affect the results, a plot of the singles count rate versus mean neutron energy were created for both the JRC and ORNL “empty” simulation results. The results of this plot, seen in Fig. 20 and 21, show that for the spectra used in this research there is not a strong correlation between mean neutron energy of a spectrum and the singles

count rate for an empty AWCC style detector. The squared Pearson correlation coefficient (R^2) is 0.40 for the JRC simulations and 0.45 for the ORNL simulations. Fig. 20 and 21 show similar trends confirming that although the AWCC and LV-AWCC differ in detailed design the underlying neutronics performance is similar. Note the trends are not simple because the mean energy is a rather crude estimate of the properties of the full spectral distribution and its interaction with the materials in the source.

Fig. 20. Comparison of the JRC singles simulation count rate results from the “empty” detector versus mean neutron energy for the simulated neutron spectra.

Fig. 21. Comparison of the ORNL singles simulation count rate results from the “empty” detector versus mean neutron energy for the simulated neutron spectra.

Energy group simulations

The results from the energy group simulations, given in Figs. 22-24, show that Am-Li neutrons with energies between 0.45 and 1,000 keV are approximately equally likely to induce fission in HEU resulting in a correlated neutron event, a double or triple count. However, this is not true for Am-Li neutrons with energies below 0.45 keV. Doubles and triples count rates appear to be roughly proportional to the logarithm of the neutron's energy. For the singles count rate, it can be seen that over the full range of energies, 0 to 1,000 keV, there is a consistent increase in counts with increasing energy. In actuality the intensity and shape of the Am-Li spectrum is poorly known at low energies, below ~10 keV, due to experimental challenges.

Fig. 22. Singles count rates MCNPX energy group simulations using the 90% HEU JRC AWCC model, with 1 sigma uncertainties. Logarithmic energy scale.

Fig. 23. Doubles count rates MCNPX energy group simulations using the 90% HEU JRC AWCC model, with 1 sigma uncertainties. Logarithmic energy scale.

Fig. 24. Triples count rates MCNPX energy group simulations using the 90% HEU JRC AWCC model, with 1 sigma uncertainties. Logarithmic energy scale.

Two main conclusions can be taken from these results. First, the doubles and triples count rates are more sensitive to changes in neutron energies below 0.45 keV in the Am-Li spectrum. However, this high sensitivity is not very important due to the fact that the majority of Am-Li neutrons are created with energies above 1 keV. Second, there appears to be an optimal neutron interrogation energy where the singles count rate is significantly reduced without significantly impacting the doubles and triples count rates. This effectively reduces the uncertainty in the doubles count rate, thus improving the capability of the detector. This optimal neutron interrogation energy is examined in more detail below.

The doubles count rate uncertainty can be estimated from expected count rate values and measurement parameters [33]. Equations 1 – 4 show these relationships:

$$\sigma_D = \xi_D \sqrt{(D + 2S^2 T_G)/t} \quad (1)$$

$$\xi_D = \sqrt{1 + 8g_2 \frac{D/f_2}{S}} \quad (2)$$

$$f_2 = e^{-T_P/\tau} (1 - e^{-T_G/\tau}) \quad (3)$$

$$g_2 = 1 - \frac{(1 - e^{-T_G/\tau})}{T_G/\tau} \quad (4)$$

where σ_D is the doubles count rate standard deviation, t is the measurement time, T_G is the gate width, T_p is the pre-delay width, and τ is the detector die-away time.

Using the JRC measurement parameters ($t = 3600$ s, $T_G = 64$ μ s, $T_p = 4.5$ μ s, and $\tau = 51$ μ s), the expected relative doubles count rate uncertainty was determined for each Am-Li neutron energy group, shown in Fig. 25. These values were then used to determine which neutron energy group produces optimal results. From Fig. 25, it can be seen that the optimal neutron energies are those below 1 eV. This makes sense since neutrons with these energies are likely to cause fission in ^{235}U (resulting in doubles counts) while easily being absorbed in the polyethylene and cadmium liner if they leave the measurement cavity (decreasing the singles counts, and thus doubles count uncertainty). While these results indicate that interrogation neutrons should have energies below 1 eV, it should be noted that this simple analysis does not capture the full complexity of active well type multiplicity measurements, which include such criteria as increasing the homogeneity of the fission rate in the measurement sample, physical constraints on which neutron energies can be created through (α ,n) reactions, and physical constraints on the size of the detector. There is currently no practical way of generating 1 eV neutrons for material assays, which limits the application of this analysis to the theoretical and simulation areas. Additionally, optimising a design for one set of items may limit the applications of a calibration / assay configuration to a broader range of items. Given the considerable effort needed to characterize and calibrate a given configuration, it is desirable not to have too many cases, but rather a few general purpose set-ups.

Fig. 25. Relative doubles count rate uncertainty values (σ_D/D) for different energy group simulations using the 90% HEU JRC AWCC model. Logarithmic energy scale.

Conclusions

Simulations using fourteen different Am-Li neutron spectra were made on two similar but different well type neutron multiplicity counters. Uranium standards of varying enrichments were measured. Although the measurement uncertainties for the doubles and triples count rates are large, the results of the fourteen simulations matched the measured results mostly within two standard deviations.

The doubles and triples count rates among the simulations compared well with each other. This is surprising due to the wide variety of spectra which include a Pu-Li spectrum from H. Ing et. al., a combined Am-Li Am-O spectrum from P. Rinard, a coarse 250 keV neutron bin width approximation from SOURCES4A, and a simplistic mono-energetic 500 keV spectrum. These results imply that the doubles and triples count rates obtained from simulations of active measurements of uranium using well type detectors are relatively insensitive to the Am-Li spectrum used. This is an important conclusion to note when comparing simulations of similar measurement types which used different Am-Li spectra.

The singles count rates on the other hand show considerable differences among the different spectra and in comparison to the physical measurements implying poor absolute agreement. Differences between the simulations is caused by the Am-Li spectra shape and mean energy, while the main potential causes of the poor agreement between the physical measurements and the simulations includes imprecise modelling; in particular, the non-detection regions at the ends of the ^3He tubes. Due to the Am-Li source locations at the end of the detector, slight changes in the ^3He non-detection regions can have a significant impact in the singles count rate. Other factors which can affect the agreement between the physical measurements and the simulations include uncertainty in the Am-Li source strength, uncertainties in nuclear data, sample position, density of the HDPE, thickness of the cadmium liner, and moisture in the detector system. Because of these uncertainties, absolute agreement between the physical measurements and the simulations should not be solely used to select which Am-Li spectra is “best”.

The suspected reason why the doubles and triples count rates are relatively insensitive to the Am-Li spectrum used while the singles count rates are not, is likely due to the source of these detected neutrons. Nearly all the neutrons which contribute to doubles and triples events originate from fission in ^{235}U induced by a neutron with epithermal energy [34, 35]. This is not true for the singles counts, which are primarily caused by Am-Li neutrons directly from the end plugs of the detector and which decrease with decreasing Am-Li neutron energy. The doubles and triples count rate uncertainties of AWCC detectors measuring HEU could be improved if the neutron energies from the interrogation sources could be reduced to values of a few tenths of an electron volt. However, doing so at this time is not viable from a practical standpoint.

In order to determine which Am-Li spectra “best” represents the true Am-Li spectrum more measurements and simulations are needed. These future measurements should consist of measuring the singles count rate from an Am-Li source with a well-known (low uncertainty) neutron emission rate. The measurement geometry should be easily reproducible with high-fidelity in simulations. The sensitivity to less well known values, like the non-detection region at the ends of the ^3He tubes, should be minimized. By using different thicknesses of moderator in a slab geometry it may be possible to statistically determine which Am-Li spectrum is most accurate. Special care should be taken to limit neutron counts due to room return. Another important aspect is to assess the content and impact of low atomic mass impurities in the Am-Li source, such as beryllium, boron, and fluorine. The contribution of Am-O neutron should also be accounted for in the simulations when not included in the reported Am-Li spectra. From a practical perspective, this work has shown that reliable predictions of performance can be made using Monte Carlo simulations when they are relative (not absolute) to a calibration. Furthermore, the repertoire of spectra assembled may be used empirically to bound a result.

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