Variations in AmLi Source Spectra and Their Estimation Utilizing the 5 Ring Multiplicity Counter

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Abstract

Active-mode assay systems are widely used for the safeguards of uranium items to verify compliance with the Non-Proliferation Treaty. Systems such as the Active-Well Coincidence Counter (AWCC) and the Uranium Neutron Coincidence Collar (UNCL) use americium-lithium (AmLi) neutron sources to induce fissions which are measured to determine the sample mass. These systems have historically relied on calibrations derived from well-defined standards. Recently, restricted access to standards or more difficult measurements have resulted in a reliance on modeling and simulation for the calibration of systems, which introduces potential simulation biases. The AmLi source energy spectra commonly used in the safeguards community do not accurately represent measurement results and the spectrum uncertainty can represent a large contribution to the total modeling uncertainty in active-mode systems.

The 5-Ring Multiplicity Counter (5RMC) has been used to measure 17 AmLi sources. The measurements showed a significant spectral variation between different sources. Utilization of a spectrum that is specific to an individual source or a series of sources will give improved results over historical general spectra when modeling AmLi sources. Candidate AmLi neutron spectra were calculated in MCNP and SOURCES4C for a range of physical AmLi characteristics. The measurement and simulation data were used to fit reliable and accurate AmLi spectra for use in the simulation of active-mode systems. Spectra were created for average Gammatron C, Gammatron N, and MRC series sources, and for individual sources. The systematic uncertainty introduced by physical aspects of the AmLi source were characterized through simulations. The accuracy of spectra from the literature was compared.

1.Introduction

Active-Mode assay systems are widely used for the safeguards verification of uranium items. Verification of uranium items represents approximately 75% of the measurements performed in IAEA inspections. Many of these verifications are performed using active-mode nondestructive assay techniques. Examples of these systems include the Active-Well Coincidence Counter (AWCC)[1] and the Uranium Neutron Coincidence Collar (UNCL)[2]. Americium-lithium (AmLi) neutron sources are used to induce fissions in samples and the resulting fissions are quantified to determine the sample mass[3]. These systems have historically relied on calibrations derived from well-defined standards. As time has progressed, access to historical standards has become increasingly difficult. In addition, the safeguards community is increasingly asked to perform active-mode system calibrations for the assay of materials for which it is not possible to manufacture or acquire representative standards. These realities have resulted in an increased reliance on system calibrations based on modeling and simulation. The sole reliance on simulation for system calibrations introduces potential simulation biases in the derived calibrations.

A critical component in the simulation bias of active-mode systems is the accurate representation of the AmLi source energy spectrum. The spectrum uncertainty can now represent a large contribution to the total modeling uncertainty in active-mode systems. Several AmLi spectra have been proposed through measurements [4-12] or calculations [13-15]. A select number of these spectra are shown in Figure 1 to highlight some key features. The SOURCES4C code is a widely trusted (α,n) simulation code. A typical AmLi spectra was simulated in SOURCES4C and included in blue in the figure for comparison. The spectra typically consist of a low energy portion from lithium and a high energy tail from oxygen (α,n) reactions. The GVZ [13] spectrum only includes the lithium portion. The high energy tail is especially important in uranium interrogation because U-238 fission can contribute at these energies. Typically these studies have only measured one source, but as we shall show, differences between sources can be important at the level of the accuracy of modern Monte Carlo simulations. However the tools and sources required to identify a manufacturer-specific or individual AmLi spectrum able to accurately reproduce measurements have not been available until now.

For this work the 5 Ring Multiplicity Counter (5RMC) was used [16]. The 5RMC measures the singles rates of the individual rings, which are dependent on the neutron energy. Six californium and seventeen AmLi sources, three Gammatron C sources, eight Gammatron N sources, and six Monsanto Research Corporation (MRC) sources, were measured as the most comprehensive study of AmLi sources to date. The large number of sources allowed the variation between sources to be characterized. An MCNP6 (version 6.1.1b) [17] model of the 5RMC was used to simulate the experiment. The procedure was to generate the possible range of lithium and oxygen (α,n) spectra based on variations of the source characteristics. The range was extended to account for unknown effects. Then least-squares fitting was used to match a combination of the spectrum from lithium or oxygen (α,n) reactions to the measurement results. There are too many unknowns for a purely analytical solution, so the measurement fitting acts as a calibration to reach a spectrum that generates an accurate detector response. Then the MCNP model was used to characterize the effects of the AmLi matrix composition, source encapsulation, and previously published spectra.

2. AmLi source effects

There are many physics considerations for the generation of neutrons in AmLi sources. To complicate things further, the determining physical characteristics of the source are poorly documented and vary between and within manufacturers. Source manufacturing was decades ago and the manufacturing companies no longer exist. Below is a summary of the issues and unknowns.

The source is typically considered as a matrix of AmO₂ particles surrounded by lithium particles. The alpha particles are generated in the AmO₂ particle and lose energy as they traverse it to reach the lithium. The alpha particles can only lose about 1 MeV before they are below the Li(α,n) threshold and can then only generate O(α,n) neutrons. The ‘infinite radius’ of the AmO₂ particle is about 10 microns, after which an increase in size does not affect the emergent alpha spectrum but increases the fraction of O(α,n) neutrons. This restriction and the particle size range of a ball mill which is commonly used in the manufacturing process, introduce a variation in the average lithium and oxygen neutron energy, and also the relative contribution by lithium and oxygen. The level of mixing has a similar effect, where two AmO₂ particles moved next to each other have less alpha particles reach lithium, but generate the same number of O(α,n) neutrons.

The form of the lithium particles is also unknown with poor documentation. Some of the compositions referenced in the literature are LiH, Li₂O, and LiOH. The composition and density of the matrix affects the relative neutron generation by lithium and oxygen. In addition, the presence of

hydrogen would thermalize the neutrons as they pass through the matrix. The manufacturer documentation frequently cites lithium mass, but it is unlikely that the matrix is pure lithium and a distinction is not made between the mass of just lithium and the mass of whatever the lithium matrix is [20]. As an example, the active dimensions of the MRC-95 source is 14.2mm length by 20.6mm diameter with an ‘isotope weight’ of 0.185 grams and ‘target weight’ of 2.3 grams. Some studies found up to 0.5% of the neutrons coming from beryllium contamination [5], although in this work gamma measurements did not indicate any beryllium contamination and so it was not considered. This can occur when other types of sources are manufactured in the same glove box, leaving behind boron or beryllium contamination that is picked up in the next AmLi source to be manufactured. These unknowns may also vary from source to source.

The Am-241 alpha particle decay energies and (α ,n) threshold energies for pure, thick target lithium, oxygen, and beryllium isotopes are listed in Table 1. The average neutron energy is from (α ,n) reactions with Am-241 alpha particles in a matrix of the element with natural isotopics, and was calculated with SOURCES4C. The alpha particle energies are below the Li-6 and O-16 thresholds, and close to the Li-7 threshold.

3.Measurements

The 5RMC is a 5 ring multiplicity counter which records the neutron rates from each individual ring. The He-3 tubes are in a polyethylene matrix, so higher energy neutrons are more likely to reach the outer rings than lower energy neutrons. Taking the ring ratios, an individual ring singles rate divided by the entire detector singles rate, normalizes the results by total emission rate to remove uncertainties introduced by the source emission strength.

Californium sources were measured due to their consistent and well-known energy spectrum. The measurements were used as a benchmark for a good spectral agreement and for the 5RMC simulations. Six californium-252 sources were measured in the 5RMC, three A7-series and three CF-series. Three Gammatron C, eight Gammatron N, and six MRC AmLi sources were also measured. Ring ratios were collected for individual sources and were also averaged for the three series.

A relative-weighted error-weighted chi squared test was used to quantify agreement between the test ring ratios and the expected ring ratios for all five rings. The test ring ratios are either the measured or simulated ring ratios being tested for agreement. The expected ring ratio is typically the averaged measured ring ratio, for example, of the Gammatron C-series. The metric is divided by the relative test ratio so that each ring is weighted equally. It is also weighted by the test and expected counting statistics, all relative to the inner ring ratio. Equation 1 shows how the chi-squared test was performed where t is test and e is expected.

$$\chi^2 = \sum_{r=1}^5 \left(\frac{test - expected}{relative_t * error_t * error_e} \right)_r^2 \quad (1)$$

4.Simulations

The MCNP model was created to match the physical detector as closely as possible. The polyethylene density is known to about 1%, so it was adjusted within this range until the 5RMC model most closely matched the measured Cf-252 ring ratios. The 5RMC response as a function of neutron energy was calculated. The response function was evaluated for on neutrons generated in a Gammatron or

MRC capsule with matrix materials of Li_2O , LiH , or LiOH , of varying densities. A material of LiOH with a density of 1.6 g cc^{-3} was found to best fit the data and was used to create the final spectra.

The neutron spectrum from (α, n) reactions in lithium and oxygen was calculated. First, alpha particles with Am-241 decay energies were transported through AmO_2 particles with radii from 1-7 microns to calculate the emergent alpha particle energy. Then these energies were used to calculate neutron spectra from (α, n) reactions in lithium and oxygen with the SOURCES4C code. The minimum and maximum values of these spectra were the bounds for the lithium and oxygen spectra fitting, extended by a factor of 4 in each direction to account for unknown variations and increase the fitting precision.

The shape of the lithium spectrum and oxygen spectrum were independently adjusted within their respective bounds. Then the two spectra were combined to form the total AmLi spectrum by adjusting the relative intensity of each. The total spectrum, when passed through the 5RMC response function, yields ring ratios. The adjustments were continued iteratively until the spectrum generated ring ratios matched the measured ring ratios. The chi-squared agreement was similar to the agreement within the Cf-252 sources. This was done for the C-series, N-series, MRC-series, and N-437 source ring ratios, generating spectra for each.

The final result is spectra generated for the three AmLi series and an individual source, based on alpha particles with degraded energy from transport through AmO_2 generating some ratio of lithium and oxygen (α, n) neutrons passed through an AmLi matrix. While individually the matrix composition, AmO_2 particle size, relative lithium contribution, and lithium and oxygen spectra, may be incorrect, the spectra that best fits the measured data on average includes the effects of these unknowns.

5.Results

The measured inner and outer ring ratios for californium are shown in Figs. 2-3, and for the AmLi sources in Figs. 4-5 respectively. The black lines are the average values and show a significant difference between the AmLi C-series and N-series sources. It can also be seen that sources with similar series numbers tend to more closely agree. This fact suggests that sources made at different times or from different 'batches' will have different spectra even within the same manufacturer. The outer ring ratios have the largest relative statistical uncertainty which is included in the figures. The statistical uncertainty for the AmLi sources is clearly negligible even in the outer ring. The measured averaged AmLi ratios for all the rings for each series are shown in Table 2.

Table 3 includes uncertainties due to counting statistics, the standard deviation between the sources, and the chi-squared agreements of each series. The standard deviation of the californium sources is nearly indistinguishable from the counting statistics, whereas the AmLi standard deviation is 5 times larger than that of californium and significantly larger than the AmLi counting uncertainty. The chi-squared value is the agreement between the worst agreeing source and the average of the series. The chi-squared agreement for Cf-252 represents the variation between Cf-252 and is the target agreement for the 5RMC MCNP model and the generated AmLi spectra. The AmLi chi-squared is larger, showing a significant variation between individual sources.

The minimum and maximum calculated lithium (α, n) neutron energies are plotted in Figure 6 as solid lines. The energies were expanded which are plotted as the dashed lines. The spectra were fit within the factor of 4 expanded bounds of the lithium and oxygen spectra and by adjusting the relative contribution between the two to minimize the chi-squared value. The change between adjacent bins was limited to force a smoother spectrum.

The results for the C-series average, N-series average, MRC-series average, and N-437 source spectra are plotted in Figs. 7a-d, and are tabulated in Appendix A. The chi-squared values and relative lithium contribution by number of neutrons are in Table 4. The N-437 source had the smallest inner ring ratio and was the hardest spectrum to fit. The lithium and oxygen energy bounds had to be expanded to a factor of 5 with no restriction on the rate of change between adjacent bins to agree to the level of the Cf-252 agreement. These spectra are emergent from the (α ,n) interactions. In practical use they should be modeled as a neutron source in a volume of 1 atom of AmO₂ to 263 atoms of LiOH at 1.6g cc⁻³ in the source encapsulation.

To show the effects of limiting the rate of change between bins in the fitting calculations, spectra were fit without this restriction. The spectra are shown in Figs. 8a-c with the agreement and lithium contribution in Table 5 and the spectra tabulated in Appendix A. It can be seen that the spectra have a more abrupt lithium peaking behavior and a less intense drop between the lithium and oxygen components. As the fitting is given more freedom it increases the intensity of the spectrum in the lithium peak, suggesting that the optimal mathematical solution approaches a monoenergetic spectrum of the lithium portion at that energy. Limiting the rate of change between bins restricts this unnatural feature. Both sets of spectra agree with the AmLi 5RMC measurements within the Cf-252 variation and are solutions to the problem.

In using MCNP to simulate the 5RMC measurements with californium the measurements and simulations do not agree within the variation between californium sources, so the MCNP model has some unknown bias. The effects of this bias should be considered when using these spectra in models of other detectors. The simulation does agree within the variation between AmLi sources. The chi-squared test results are included in Table 6, along with the model's sensitivity to polyethylene density to estimate the final uncertainty in the spectrum generation.

The average emergent alpha particle energy calculated in MCNP and corresponding average lithium neutron energy calculated in SOURCES4C as a function of AmO₂ particle size is shown in Table 7. It demonstrates the magnitude of energy loss and, when the (α ,n) threshold energies are considered, highlights how particle size can affect the AmLi neutron spectrum. The average energy reaches an asymptote as the alpha particle kinetic energy distribution approaches the 'infinite thickness' distribution, where alpha particles from an increased radius don't have the range to escape the particle.

The matrix material that best agreed with the data was LiOH with a density of 1.6 g/cc. This and other material's plausibility is discussed in other work [21]. Figure 9 shows the simulated 5RMC response function for the measurement conditions, the LiOH matrix material in Gammatron model AN-HP-9 encapsulation in a tungsten pig. The effects of changing individual aspects of the simulation are shown in Table 8 by keeping the source spectrum the same. The first entry is the baseline simulation of as-measured conditions. The matrix materials were changed in the next four entries. As a calculation check, the next entry is the spectrum input directly into MCNP instead of calculated with the response function, which should closely match the baseline entry and shows the uncertainty introduced through the response function calculation. 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 AmLi sources have to reduce gamma emissions. Removing the 423 grams of tungsten demonstrates the sensitivity of the chi-squared metric.

To compare the effectiveness of the generated spectra, historical spectra were simulated to calculate their ring ratios. Their chi-squared agreements with the C-series, N-series, MRC-series, and N-437 source ratios are shown in Table 9. The closest agreements are bolded. The first three spectra are this

work. The following spectra are measured and include interaction with the AmLi matrix and the encapsulation. The spectra below Werle are initial spectra before interaction with the AmLi matrix or encapsulation. The final spectra is from simulating in SOURCES4C the AmLi material chosen for this work, 1 atom of AmO₂ to 263 atoms of LiOH with a density of 1.6 g cc⁻³. In SOURCES4C the Am atoms are evenly distributed in an infinite matrix of O, Li, and H atoms, there are no particles simulated. The agreement of that spectrum is the worst of all the agreements, suggesting that this simplification introduces significant errors.

6. Conclusions

This work comprises three main results. First, measurements were used to characterize the variations in AmLi and Cf-252 sources, and 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 simulations of AmLi sources, reducing the spectrum-dependent systematic uncertainty. Finally, the simulations were used to demonstrate the sensitivity to various aspects of the system, including the detector polyethylene density, the lithium compound composition, the source encapsulation, and historical measured and analytically calculated spectra from the literature. The results show that a change in any one of these parameters is enough to change the spectrum more than the variation between Cf-252 sources. A variation of a combination of these factors is the likely reason for large differences within the many AmLi spectra. The limited set of measurements in this work suggest an even larger variation in AmLi sources across the world.

While this was the most comprehensive study of the variation in AmLi spectra to date, the comparison metric was limited to terms of agreement within the variation of measured Cf-252 sources. This is scientifically meaningful but is of limited use to a specific active assay system. As an extension the approach demonstrated here should be applied to a practical system such as the Uranium Neutron Coincidence Collar to understand the improvement in accuracy of assay of U-235 mass. An effort should be made to calibrate such a system using a specific source's generated spectra and confirm with measurements using that source. The ultimate objective would be AmLi spectra accurate enough to enable simulated calibrations, adding significant capabilities to active interrogation systems.

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Figure 1. Selected AmLi spectra normalized per bin width.

Figure 2. Cf-252 inner ring ratios and associated statistical uncertainty.

Figure 3. Cf-252 outer ring ratios and associated statistical uncertainty.

Figure 4. AmLi inner ring ratios and associated statistical uncertainty.

Figure 5. AmLi outer ring ratios and associated statistical uncertainty.

Figure 6. Lithium (α,n) spectrum bounds

Figures 7a, 7b, 7c, and 7d. AmLi Gammatron C-series, N-series, MRC-series and N-437 source generated spectrum

Figures 8a, 8b, and 8c. AmLi C-series, N-series, and MRC-series generated spectra without adjacent bin rate of change restrictions.

Figure 9. 5RMC with LiOH matrix and Gammatron capsule response function.

Figure 9

Figure 9: γ UW

Figure 10

Figure 1

Figure 10

Figure 1 VUW

Figure 10: Cf-252

Figure 1 (continued)

Figure 1 (continued)

Figure 9. Weighted

Figure 10

Figure 4U

Figure 1V

Figure 1W

Figure +X

Figure 1. U

Figure 1, V

Figure 1, W

Figure - VUW

Figure - Wcf

Table 1. Physical properties of the relevant isotopes [18-19]

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

Table 2. C, N, and MRC series measured ring ratios

Ratio	C-series	N-series	MRC-series
R1	0.49541	0.49016	0.48964
R2	0.35387	0.35472	0.35548
R3	0.11465	0.11704	0.11733
R4	0.02891	0.03027	0.03007
R5	0.00714	0.00779	0.00747

Table 3. Agreement values of the Cf-252 and AmLi inner ring ratio measurements

	Cf-252	AmLi
Statistical uncertainty	0.11%	0.01%
Standard deviation	0.12%	0.55%
Chi-squared (C,N,MRC)	1.4×10^{-6}	1.7×10^{-5} , 6.2×10^{-5} , 4.1×10^{-6}

Table 4. Chi-squared values and lithium contribution percent for the generated AmLi spectra

Source	Chi-squared	Lithium contribution (%)
C-series	1.3×10^{-6}	93.3%
N-series	1.4×10^{-6}	91.9%
MRC-series	1.1×10^{-6}	93.3%
N-437	1.4×10^{-6}	92.1%

Table 5. Chi-squared values and lithium contribution percent for the generated spectra without slope restrictions.

Source	Chi-squared	Lithium contribution (%)
C-series	1.0×10^{-6}	92.8%
N-series	1.3×10^{-6}	91.8%
MRC-series	9.3×10^{-7}	92.8%

Table 6. Chi-squared agreements of the 5RMC model with Cf-252

Simulation	Chi-squared value
5RMC best MCNP model agreement with average Cf-252 sources (poly $\rho=0.953 \text{ g cc}^{-1}$)	$9.0 \cdot 10^{-6}$
5RMC MCNP model agreement (poly $\rho=0.950 \text{ g cc}^{-1}$)	$1.7 \cdot 10^{-5}$

Table 7. Alpha and neutron average energies as a function of particle size

Particle radius (micrometer)	Emergent average alpha energy (MeV)	Li (α,n) average neutron energy (MeV)	O (α,n) average neutron energy (MeV)
0.5	5.36	0.560	2.43
1	5.22	0.528	2.39
3	4.68	0.501	2.29
5	4.09	0.495	2.25
7	3.39	0.494	2.24
9	2.79	0.494	2.24

Table 8. Chi-squared agreement between the C-series sources and the generated spectrum for different matrices

Matrix, density (g cc^{-1})	Chi-squared
LiOH, 1.6 (calculated from response function)	$1.3 \cdot 10^{-6}$
LiOH, 1.32	$2.0 \cdot 10^{-5}$
Li ₂ O, 0.75	$4.8 \cdot 10^{-4}$
LiH, 0.56	$1.6 \cdot 10^{-5}$
LiH, 1.0	$1.4 \cdot 10^{-4}$
LiOH, 1.6 (calculated directly in MCNP)	$1.2 \cdot 10^{-6}$
LiOH, 1.6 (MRC encapsulation)	$1.8 \cdot 10^{-5}$
LiOH, 1.6 (Gammatron encapsulation, no tungsten pig)	$2.1 \cdot 10^{-6}$

Table 9. Historical spectra simulated in the 5RMC MCNP model agreement with the measured results

Spectra	Chi-Squared			
	C-series	N-series	MRC-series	N-437
Generated C-series	$1.3 \cdot 10^{-6}$	$4.9 \cdot 10^{-5}$	$5.8 \cdot 10^{-5}$	$9.5 \cdot 10^{-5}$
Generated N-series	$4.8 \cdot 10^{-5}$	$1.4 \cdot 10^{-6}$	$3.0 \cdot 10^{-6}$	$9.9 \cdot 10^{-6}$
Generated MRC-series	$5.7 \cdot 10^{-5}$	$2.6 \cdot 10^{-6}$	$1.1 \cdot 10^{-6}$	$6.0 \cdot 10^{-6}$
Generated N-437	$9.4 \cdot 10^{-5}$	$9.4 \cdot 10^{-6}$	$6.1 \cdot 10^{-6}$	$1.4 \cdot 10^{-6}$
Birch [8]	$3.4 \cdot 10^{-5}$	$1.3 \cdot 10^{-4}$	$1.5 \cdot 10^{-4}$	$2.0 \cdot 10^{-4}$
Delafield [9]	$4.7 \cdot 10^{-5}$	$1.1 \cdot 10^{-5}$	$1.5 \cdot 10^{-5}$	$2.5 \cdot 10^{-5}$
Ing [10]	$4.9 \cdot 10^{-5}$	$2.5 \cdot 10^{-5}$	$3.6 \cdot 10^{-5}$	$4.9 \cdot 10^{-5}$
Obninsk [6]	$2.4 \cdot 10^{-5}$	$6.0 \cdot 10^{-5}$	$7.7 \cdot 10^{-5}$	$1.9 \cdot 10^{-4}$
Owen0.1 [11]	$4.2 \cdot 10^{-4}$	$1.9 \cdot 10^{-4}$	$1.8 \cdot 10^{-4}$	$1.3 \cdot 10^{-4}$
Owen5 [11]	$6.5 \cdot 10^{-4}$	$3.6 \cdot 10^{-4}$	$3.5 \cdot 10^{-4}$	$2.8 \cdot 10^{-4}$

Tagziria 2003 [4]	$1.0 \cdot 10^{-4}$	$1.9 \cdot 10^{-4}$	$2.2 \cdot 10^{-4}$	$2.6 \cdot 10^{-4}$
Tagziria 2004 [5]	$8.3 \cdot 10^{-4}$	$4.9 \cdot 10^{-4}$	$4.6 \cdot 10^{-4}$	$3.7 \cdot 10^{-4}$
Werle [12]	$5.3 \cdot 10^{-5}$	$1.9 \cdot 10^{-4}$	$2.1 \cdot 10^{-4}$	$2.8 \cdot 10^{-4}$
Geiger [13]	$4.6 \cdot 10^{-5}$	$2.3 \cdot 10^{-5}$	$3.2 \cdot 10^{-5}$	$4.6 \cdot 10^{-5}$
Tagziria 2012 [14]	$9.5 \cdot 10^{-5}$	$2.5 \cdot 10^{-5}$	$3.5 \cdot 10^{-5}$	$3.5 \cdot 10^{-5}$
Sources99 [15]	$3.1 \cdot 10^{-4}$	$1.3 \cdot 10^{-4}$	$1.3 \cdot 10^{-4}$	$9.2 \cdot 10^{-5}$
SOURCES4C	$1.1 \cdot 10^{-2}$	$9.4 \cdot 10^{-3}$	$9.4 \cdot 10^{-3}$	$9.0 \cdot 10^{-3}$

Appendix A: Spectra tabulated

Energy is the upper bin limit in MeV. The intensities are not normalized by bin width.

Energy (MeV)	C-series	N-series	MRC- series	N-437	C-series unrestricted	N-series unrestricted	MRC- series unrestricted
0.025	2.1911E-03	1.1266E-03	2.2284E-03	1.0036E-03	1.0280E-03	1.2258E-03	1.2927E-03
0.05	3.4530E-03	2.0392E-03	4.6726E-03	1.7878E-03	1.8596E-03	2.2195E-03	2.3738E-03
0.075	5.2562E-03	2.7376E-03	7.0621E-03	2.3744E-03	2.4753E-03	2.9521E-03	3.1611E-03
0.1	7.3454E-03	3.5354E-03	9.6859E-03	2.9600E-03	3.0599E-03	3.6402E-03	3.8615E-03
0.15	1.9239E-02	8.1421E-03	2.4938E-02	6.0314E-03	6.3147E-03	7.4902E-03	8.0129E-03
0.2	2.5086E-02	1.0342E-02	3.1604E-02	6.0555E-03	6.3367E-03	7.5270E-03	8.0580E-03
0.25	1.6822E-02	7.8949E-03	2.0451E-02	3.0481E-03	4.1881E-02	3.7850E-03	6.7202E-02
0.275	2.3146E-02	1.4603E-02	2.6696E-02	3.0467E-03	3.1669E-03	3.7826E-03	6.2962E-02
0.3	3.2494E-02	3.1454E-02	3.5848E-02	3.0405E-03	8.0339E-02	3.7744E-03	6.6756E-02
0.325	4.6296E-02	7.3754E-02	4.8774E-02	3.0291E-03	7.9824E-02	9.5847E-02	6.6328E-02
0.35	6.6592E-02	8.7241E-02	6.6951E-02	1.0876E-01	7.9099E-02	9.4976E-02	6.5725E-02
0.375	9.6389E-02	8.6246E-02	9.2473E-02	1.0752E-01	7.8197E-02	9.3893E-02	6.4976E-02
0.4	9.9088E-02	8.5057E-02	1.2820E-01	1.0603E-01	7.7120E-02	9.2599E-02	6.4083E-02
0.425	9.7435E-02	8.3641E-02	8.9685E-02	1.0427E-01	7.5836E-02	9.1058E-02	6.3018E-02
0.45	6.5243E-02	8.2022E-02	6.3000E-02	1.0225E-01	7.4369E-02	8.9295E-02	6.1800E-02
0.475	9.3390E-02	8.0172E-02	4.4531E-02	9.9939E-02	7.2691E-02	8.7281E-02	6.0407E-02
0.5	6.2162E-02	7.6928E-02	3.1785E-02	9.7381E-02	7.0831E-02	8.5047E-02	5.8503E-02
0.525	4.1644E-02	7.5896E-02	2.7603E-02	9.4608E-02	6.8814E-02	8.2625E-02	5.6703E-02
0.55	2.8317E-02	3.1102E-02	3.6508E-02	3.1611E-02	6.6976E-02	2.2990E-02	3.0778E-02
0.575	1.9614E-02	1.3913E-02	2.6214E-02	2.7665E-03	2.7797E-03	3.3281E-03	5.3506E-02
0.6	1.3933E-02	7.1872E-03	1.9202E-02	2.6158E-03	2.7089E-03	3.2434E-03	6.9305E-03
0.625	1.0192E-02	9.2414E-03	1.4384E-02	2.5397E-03	2.6300E-03	3.1489E-03	3.3682E-03
0.65	7.7030E-03	5.2622E-03	1.1039E-02	2.4676E-03	2.5550E-03	3.0589E-03	3.4111E-03
0.675	6.0327E-03	3.6482E-03	8.7005E-03	2.3941E-03	2.4782E-03	2.9667E-03	3.1680E-03
0.7	4.9023E-03	2.9648E-03	7.0569E-03	2.3211E-03	2.4015E-03	2.8745E-03	3.0680E-03
0.725	4.1201E-03	3.4297E-03	5.8796E-03	2.2430E-03	2.3197E-03	2.7762E-03	2.9939E-03
0.75	3.5714E-03	2.7698E-03	5.0286E-03	2.1682E-03	2.2401E-03	2.6802E-03	2.9500E-03
0.775	3.8346E-03	2.4530E-03	4.3943E-03	2.0869E-03	2.1543E-03	2.5769E-03	2.7736E-03
0.8	3.3086E-03	2.2792E-03	3.9283E-03	2.0101E-03	2.0739E-03	2.4804E-03	3.2735E-03

0.825	2.9271E-03	2.2661E-03	3.5657E-03	1.9293E-03	1.9899E-03	2.3798E-03	2.5317E-03
0.85	2.6451E-03	2.3661E-03	3.2769E-03	1.8548E-03	1.9109E-03	2.2845E-03	2.4272E-03
0.875	2.6788E-03	2.0833E-03	3.2549E-03	1.7801E-03	1.8308E-03	2.1877E-03	2.3202E-03
0.9	2.3958E-03	1.9119E-03	3.2609E-03	1.6963E-03	1.7418E-03	2.0804E-03	2.2025E-03
0.925	2.4059E-03	1.9219E-03	3.1889E-03	1.4482E-03	1.4881E-03	1.7760E-03	2.1814E-03
0.95	2.0732E-03	1.8619E-03	2.7940E-03	1.3823E-03	1.3687E-03	1.6101E-03	2.0425E-03
0.975	2.1402E-03	1.5847E-03	2.9054E-03	1.1911E-03	1.2572E-03	1.4850E-03	1.9691E-03
1	1.7832E-03	1.7640E-03	2.4615E-03	1.1213E-03	1.1368E-03	1.3824E-03	1.7870E-03
1.025	1.8643E-03	1.2667E-03	2.4195E-03	1.0498E-03	1.1013E-03	1.2681E-03	1.4502E-03
1.05	1.5508E-03	1.3212E-03	2.1211E-03	9.2954E-04	1.0557E-03	1.1717E-03	1.3884E-03
1.075	1.5363E-03	1.5961E-03	2.0196E-03	9.3407E-04	8.9254E-04	1.0708E-03	1.3183E-03
1.1	1.3884E-03	2.2172E-03	1.6777E-03	7.6540E-04	8.0480E-04	9.6544E-04	1.0981E-03
1.125	1.2985E-03	1.5364E-03	1.7217E-03	6.7787E-04	7.1328E-04	8.5551E-04	1.1278E-03
1.15	1.2257E-03	1.0323E-03	1.5765E-03	5.8715E-04	6.7889E-04	7.4169E-04	1.0922E-03
1.175	9.7711E-04	1.0430E-03	1.3020E-03	5.6935E-04	5.1982E-04	6.2308E-04	1.0648E-03
1.2	9.8742E-04	6.9684E-04	8.6537E-04	3.9996E-04	4.2285E-04	5.0660E-04	1.0237E-03
1.225	6.9784E-04	1.0480E-03	7.1475E-04	3.4676E-04	3.7810E-04	4.3988E-04	6.8088E-04
1.25	6.9881E-04	1.3315E-03	6.0818E-04	3.0602E-04	3.2489E-04	3.8886E-04	8.2876E-04
1.275	6.3922E-04	1.3852E-03	6.3523E-04	3.2896E-04	2.8356E-04	3.3918E-04	8.6013E-04
1.3	5.4943E-04	6.6227E-04	5.3610E-04	2.2659E-04	2.4203E-04	2.8928E-04	4.5920E-04
1.325	5.1298E-04	1.1702E-03	3.7919E-04	2.6572E-04	1.9441E-04	2.3209E-04	7.7150E-04
1.35	4.4442E-04	5.9788E-04	3.0425E-04	1.4372E-04	1.5539E-04	1.8521E-04	3.0947E-04
1.375	2.9728E-04	4.8639E-04	3.3409E-04	1.1232E-04	1.2273E-04	1.4593E-04	6.9138E-04
1.4	1.1831E-04	2.0733E-04	1.3642E-04	8.4560E-05	9.3922E-05	1.1126E-04	1.2990E-04
1.425	8.4080E-05	1.5703E-04	9.4282E-05	6.0986E-05	6.9496E-05	8.1858E-05	9.8574E-05
1.45	4.8342E-05	1.2955E-04	4.7097E-05	3.5494E-05	4.3925E-05	5.0837E-05	6.7051E-05
1.475	3.7014E-05	1.3316E-04	3.2221E-05	2.6127E-05	3.3545E-05	3.8534E-05	5.2729E-05
1.5	4.4682E-05	1.7022E-04	3.8684E-05	3.1589E-05	4.0636E-05	4.6662E-05	6.3968E-05
1.6	1.5968E-04	3.5559E-04	1.5463E-04	1.2962E-04	1.6682E-04	1.9154E-04	2.6269E-04
1.7	1.7792E-04	3.9596E-04	1.7232E-04	1.4445E-04	1.8591E-04	2.1346E-04	2.9276E-04
1.8	2.0678E-04	4.4647E-04	2.0116E-04	1.6879E-04	2.1723E-04	2.4942E-04	3.4208E-04
1.9	3.3681E-03	4.0326E-03	3.4918E-03	4.2213E-03	3.8066E-03	4.3714E-03	3.9025E-03
2	6.2341E-03	7.4693E-03	6.4589E-03	7.8416E-03	7.0513E-03	8.0968E-03	7.1892E-03
2.2	7.6590E-03	9.1727E-03	7.9381E-03	9.5865E-03	8.6590E-03	9.9434E-03	8.9020E-03
2.4	8.0663E-03	9.6620E-03	8.3591E-03	1.0100E-02	9.1211E-03	1.0474E-02	9.3721E-03
2.6	7.6073E-03	9.1125E-03	7.8831E-03	9.5266E-03	8.6023E-03	9.8781E-03	8.8380E-03
2.8	6.5965E-03	7.9014E-03	6.8360E-03	8.2595E-03	7.4590E-03	8.5652E-03	7.6650E-03
3	6.0106E-03	7.1996E-03	6.2287E-03	7.5355E-03	6.7965E-03	7.8045E-03	6.9684E-03
3.25	4.8576E-03	5.8163E-03	5.0356E-03	6.0845E-03	5.4904E-03	6.3050E-03	5.6354E-03
3.5	3.0994E-03	3.7092E-03	3.2144E-03	3.8827E-03	3.5012E-03	4.0209E-03	3.5902E-03
3.75	1.6914E-03	2.0220E-03	1.7560E-03	2.1195E-03	1.9084E-03	2.1919E-03	1.9529E-03

4	7.9845E-04	9.5205E-04	8.3073E-04	1.0004E-03	8.9833E-04	1.0321E-03	9.1620E-04
4.25	3.4075E-04	4.0399E-04	3.5627E-04	4.2668E-04	3.8097E-04	4.3798E-04	3.8586E-04
4.5	1.2638E-04	1.4761E-04	1.3383E-04	1.5671E-04	1.3899E-04	1.6006E-04	1.4033E-04
4.75	6.5989E-05	7.5772E-05	7.0865E-05	8.0904E-05	7.1221E-05	8.2177E-05	7.1658E-05
5	4.5586E-05	5.1807E-05	4.9363E-05	5.5614E-05	4.8642E-05	5.6193E-05	4.8666E-05
5.25	2.1755E-05	2.6086E-05	2.2525E-05	2.7282E-05	2.4628E-05	2.8277E-05	2.5275E-05
5.5	1.0405E-05	1.2476E-05	1.0773E-05	1.3113E-05	1.1779E-05	1.3524E-05	1.1980E-05
5.75	2.2321E-06	2.6765E-06	2.3111E-06	2.8256E-06	2.5269E-06	2.9013E-06	2.5494E-06
6	2.6273E-07	3.1503E-07	2.7202E-07	3.3186E-07	2.9742E-07	3.4149E-07	3.0127E-07
6.25	2.2102E-07	2.6502E-07	2.2884E-07	2.7918E-07	2.5020E-07	2.8728E-07	2.5344E-07
6.5	1.8556E-07	2.2250E-07	1.9212E-07	2.3439E-07	2.1006E-07	2.4119E-07	2.1278E-07
6.75	1.5551E-07	1.8647E-07	1.6101E-07	1.9643E-07	1.7604E-07	2.0213E-07	1.7832E-07
7	1.3009E-07	1.5599E-07	1.3469E-07	1.6433E-07	1.4727E-07	1.6909E-07	1.4917E-07
7.25	1.0868E-07	1.3031E-07	1.1252E-07	1.3728E-07	1.2303E-07	1.4126E-07	1.2461E-07
7.5	9.0635E-08	1.0868E-07	9.3840E-08	1.1449E-07	1.0260E-07	1.1781E-07	1.0392E-07
7.75	7.5469E-08	9.0492E-08	7.8138E-08	9.5331E-08	8.5434E-08	9.8094E-08	8.6534E-08
8	6.2762E-08	7.5255E-08	6.4981E-08	7.9279E-08	7.1049E-08	8.1577E-08	7.1963E-08
8.25	5.2125E-08	6.2501E-08	5.3969E-08	6.5844E-08	5.9008E-08	6.7752E-08	5.9767E-08
8.5	4.3237E-08	5.1844E-08	4.4766E-08	5.4617E-08	4.8946E-08	5.6199E-08	4.9575E-08
8.75	3.5824E-08	4.2955E-08	3.7090E-08	4.5252E-08	4.0554E-08	4.6563E-08	4.1075E-08
9	2.9639E-08	3.5538E-08	3.0687E-08	3.7439E-08	3.3552E-08	3.8524E-08	3.3983E-08
9.25	2.4500E-08	2.9377E-08	2.5366E-08	3.0948E-08	2.7735E-08	3.1844E-08	2.8091E-08
9.5	2.0231E-08	2.4259E-08	2.0947E-08	2.5556E-08	2.2903E-08	2.6296E-08	2.3197E-08
9.75	1.6693E-08	2.0016E-08	1.7284E-08	2.1087E-08	1.8897E-08	2.1698E-08	1.9140E-08
10	1.3752E-08	1.6489E-08	1.4238E-08	1.7371E-08	1.5568E-08	1.7874E-08	1.5767E-08
10.25	1.1323E-08	1.3577E-08	1.1723E-08	1.4303E-08	1.2818E-08	1.4717E-08	1.2982E-08
10.5	9.3151E-09	1.1169E-08	9.6445E-09	1.1767E-08	1.0545E-08	1.2108E-08	1.0680E-08
10.75	7.6584E-09	9.1828E-09	7.9292E-09	9.6742E-09	8.6696E-09	9.9543E-09	8.7807E-09
11	6.2875E-09	7.5390E-09	6.5098E-09	7.9424E-09	7.1176E-09	8.1723E-09	7.2088E-09
11.25	5.1594E-09	6.1864E-09	5.3418E-09	6.5174E-09	5.8406E-09	6.7061E-09	5.9154E-09