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Simulation of the Neutron Response Matrix for a Liquid Scintillator and Spectrum Unfolding

May 2005

Prepared by
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SIMULATION OF THE NEUTRON RESPONSE MATRIX FOR A LIQUID SCINTILLATOR AND SPECTRUM UNFOLDING

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CONTENTS

	Page
LIST OF FIGURES	v
ABSTRACT	vii
1. INTRODUCTION	1
2. DETECTOR	1
3. FORIST UNFOLDING CODE	1
4. RESPONSE MATRIX	1
5. SIMULATED MATRIX	3
6. SMOOTHED MATRIX	4
7. MEASURED RESPONSE MATRIX	5
8. TEST CASES	7
8.1 Cf-252 NEUTRON SPECTRUM	7
8.2 14 MeV NEUTRONS	9
8.3 4 MeV AND 14 MeV NEUTRONS	10
8.4 Am-Be NEUTRON SPECTRUM	12
8.5 MEASUREMENT OF Cf-252 SOURCE	13
9. CONCLUSIONS AND FUTURE WORK	14
REFERENCES	15

LIST OF FIGURES

Figure		Page
1	The Illinois matrix (provided with the FORIST code).	2
2	Some response curves taken from the Illinois matrix. Each column of the matrix corresponds to a different energy. In this plot, pulse heights are converted to MeVee.	3
3	The simulated response matrix for our detector.	4
4	Some response curves taken from the simulated response matrix. Each column of the matrix corresponds to a different energy.	4
5	Some response curves taken from the modified matrix. Each column of the matrix corresponds to a different energy bin.	5
6	Measured response matrix	6
7	Some response curves taken from the measured matrix. Each column of the matrix corresponds to a different energy.	7
8	Cf spectrum unfolded with the Illinois matrix. The error bars indicate plus and minus one standard deviation, as calculated by the FORIST code	8
9	Cf spectrum unfolded with the simulated matrix.	8
10	Cf spectrum unfolded with the modified simulated matrix.	9
11	14 MeV neutrons, unfolded with the Illinois matrix.	10
12	14 MeV neutrons, unfolded with the simulated matrix.	10
13	14 MeV neutrons, unfolded with the modified simulated matrix.	11
14	4 MeV and 14 MeV neutrons, unfolded with the Illinois matrix.	11
15	4 MeV and 14 MeV neutrons, unfolded with the simulated matrix.	12
16	Am-Be neutron spectrum, unfolded with the simulated matrix.	13
17	Am-Be neutron spectrum, unfolded with the modified simulated matrix.	13
18	Measured Cf-252 neutron spectrum.	14

ABSTRACT

This paper presents the results of measurements and Monte Carlo simulations aimed at determining the response matrix of a liquid scintillation detector. The response matrix can be used in conjunction with neutron unfolding codes to determine the spectrum of the incident neutron field on the basis of the pulse height response measured by the scintillator. Knowledge of the neutron spectrum is highly desirable and cannot be obtained with the traditionally used thermal neutron detectors. The analysis presented includes Monte Carlo simulations of the response matrix, and comparisons with the measured response matrix. The simulations were performed with the MCNP-PoliMi code, which models the detector response by taking into account the light output of neutron interactions on hydrogen and carbon nuclei present in the scintillator. We also present some preliminary approaches to the neutron spectrum unfolding problem.

The unfolding was performed with the FORIST code, which is well-benchmarked and widely used. Both simulated and measured neutron pulse height spectra were unfolded. The simulations include monoenergetic neutrons, a Cf-252 source, and an Am-Be source. The measurements were performed with a Cf-252 source, and the neutrons were discriminated from the photons by a time-of-flight method. We compare the results obtained with different response matrices: the matrix provided with the code, our simulated response matrix, and our measured response matrix.

1. INTRODUCTION

Recent studies[1] have shown that the measurement of neutron spectra can increase the sensitivity of assays performed on nuclear materials. Neutron spectrum unfolding with organic scintillators requires the knowledge of the neutron response matrix, which links the neutron energy with the pulse heights.

The purpose of this work is to determine the neutron response matrix for a liquid organic scintillator and use it to perform neutron spectrum unfolding.

2. DETECTOR

The detector is a cylindrical liquid scintillator manufactured by Bicron, model 4.62MAB-1F3BC-501A/5. It is 7.7 cm thick, with a 15.2-cm diameter. This liquid scintillator is bubble-free and is surrounded by an expansion tube containing oxygen-free nitrogen. The container is made of aluminum. The front face is approximately 2 mm thick. The side is a double aluminum wall with an external layer 2 mm thick and an internal layer 0.5 mm thick. Between the two layers there is 1.5 cm of nitrogen. The inside of the container has a clear-anodized surface treatment. The photomultiplier tube is mounted on the back circular surface.

3. FORIST UNFOLDING CODE

FORIST[2] (FERDOR with Optimized Resolution using an Iterative Smoothing Technique) is a modification of the FERDOR code.[3] It is assumed[4] that the response of the detector is given by $Ax = b$, where A is the detector response function matrix, x is the unknown spectrum, and b is the pulse height distribution. The code uses a matrix inversion method to calculate a lower and upper bound for each element of the solution vector x. The solution vector x is constrained to satisfy the equation

$$Ax = b + \varepsilon$$

where ε is the stochastic counting error vector, and it is assumed that the counting errors are independent of one another and are normally distributed with mean zero.

The FORIST code is written for measurements consisting of two different gain runs. Because the response matrix used in the method includes the resolution of the spectrometer, the elements of the solution vector have large statistical errors and require smoothing. The smoothing functions are determined iteratively to optimize the energy resolution.

4. RESPONSE MATRIX

FORIST is currently dimensioned for a 113 by 81 response matrix. The 81 columns of the response matrix are response functions corresponding to different energies, from 0.2 to 22 MeV. The 113 rows correspond to 113 pulse height bins, from 0.06 to 14 MeVee, approximately.

Our first results were unfolded using the Illinois neutron response matrix,[5] which is supplied with the FORIST code. The Illinois matrix is shown in Fig. 1. This matrix is based on the measurements of Verbinski et al.[6] for a 4.60×4.65 cm liquid organic scintillator, integrated with simulations, interpolations and extrapolations. The scintillator material is NE-213, which is identical to BC 501. The pulse height is

given in light units, where one light unit is defined as the position of the half-height of the Compton edge produced in NE-213 by the 1.28 MeV Na-22 gamma rays.

The energy and pulse height binning of the Illinois matrix is irregular. Energies vary from 0.2 to 22 MeV. Energy bin widths vary from 0.08 to 0.18 MeV. Pulse heights vary from 0.05 to 12 light units (from 0.06 to 14 MeVee, approximately). Pulse height bin widths vary from 0.0125 to 0.3 light units (from 0.014 to 0.35 MeVee, approximately).

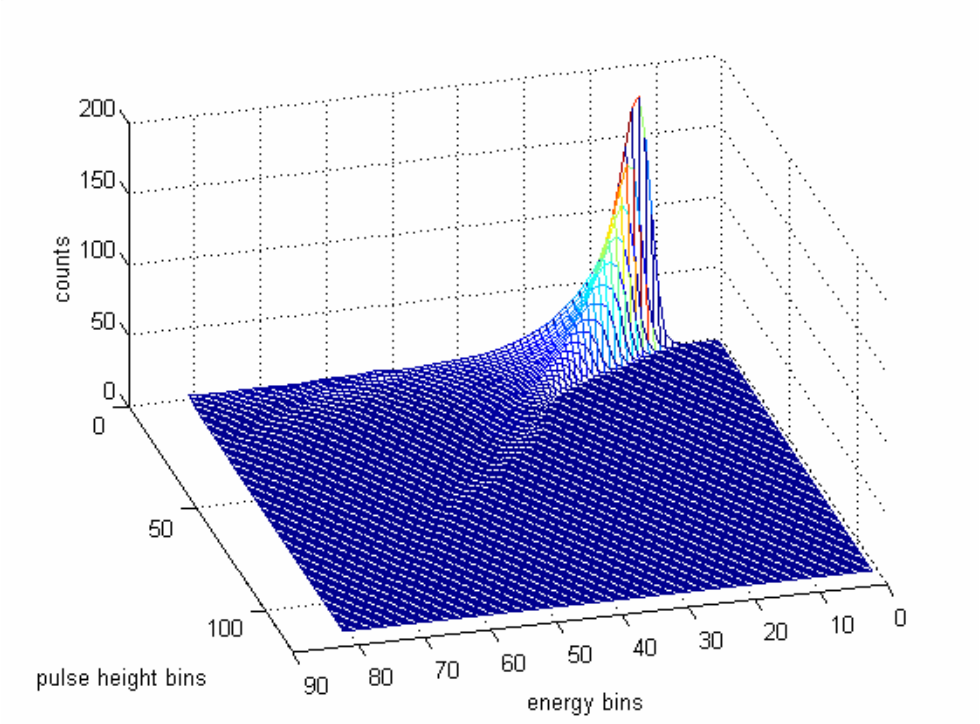

Fig. 1. The Illinois matrix (provided with the FORIST code).

Figure 2 shows some neutron response functions taken from the Illinois matrix. The functions are approximately rectangular, as expected for a neutron detector based on proton recoil[7].

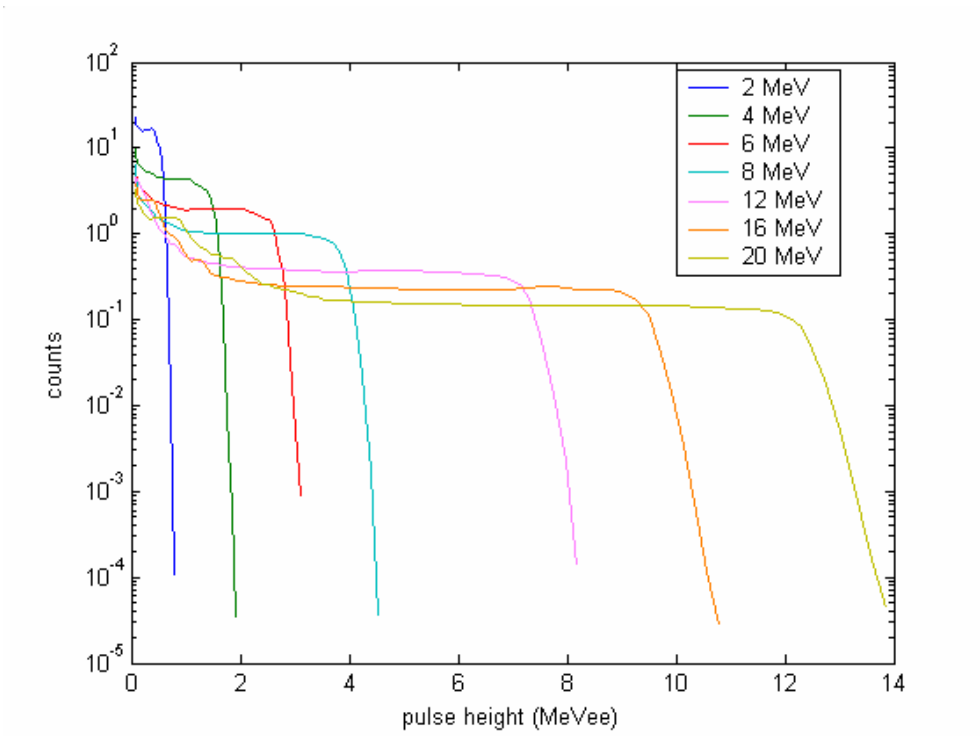

Fig. 2. Some response curves taken from the Illinois matrix. Each column of the matrix corresponds to a different energy. In this plot, pulse heights are converted from light units to MeVee.

5. SIMULATED MATRIX

The results obtained with the Illinois matrix are reasonable but not very accurate (see Section 8). In particular, in unfolding monoenergetic neutron spectra the energies obtained with that matrix are approximately 1 MeV higher or lower than the actual energies. This effect is probably due to the fact that the Illinois matrix is based on a detector that is different from the one we used. This is why it seems necessary to build a neutron response matrix that is specific for the detector in use.

One possible approach to determining the response matrix is Monte Carlo simulations. We used the MCNP-PoliMi code,[8] which gives a detailed record of each interaction occurring in the detector (particle, interaction type, energy, position, and time).

A post-processing Matlab code analyzes the interactions occurring in the scintillator material and calculates the corresponding light output. This code takes into account neutron scattering on hydrogen, neutron scattering on carbon, and secondary photons. The calibration energy deposition-light output was measured for the NE-213 liquid scintillator.[9] Previous works[10] have shown that response functions calculated with the MCNP- PoliMi code are in good agreement with measurements.

Figure 3 shows our simulated response matrix. The pulse heights were binned in an irregular binning structure analogous to that of the Illinois matrix, so that we could use the FORIST unfolding code without modifying it.

In Fig. 3 we can see a structure at high energies and low pulse height, which is not present in the Illinois matrix. This structure is due to (n,α) reactions on carbon. The post-processing code assigns a small light output to all interactions with carbon. This light output is proportional to the energy deposited.

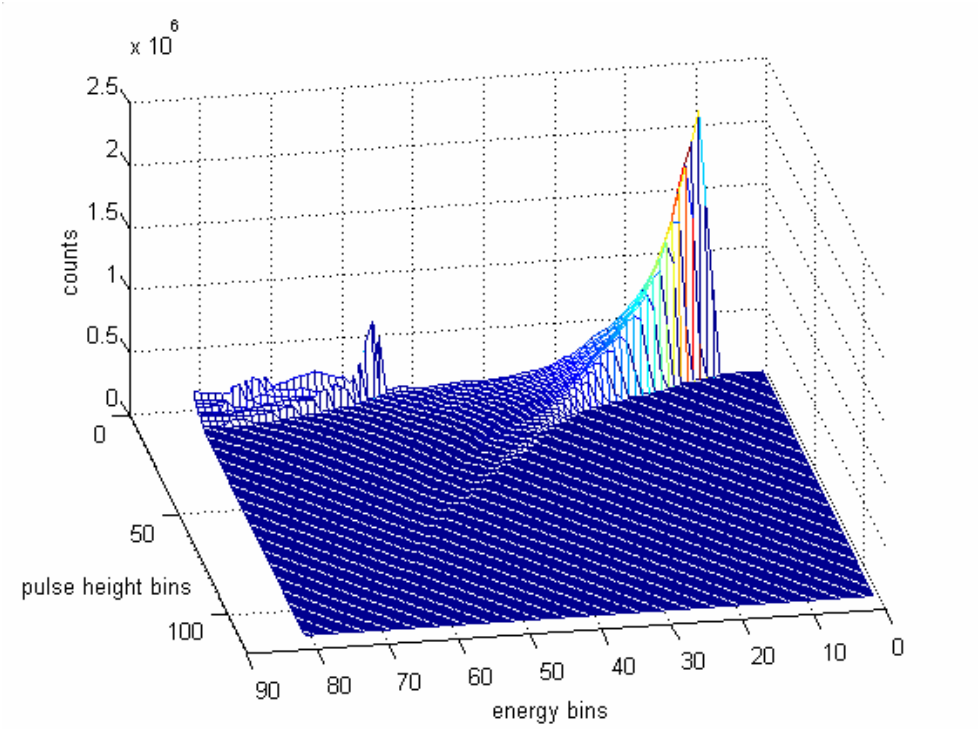

Fig. 3. The simulated response matrix for our detector.

Figure 4 shows some neutron response functions taken from the simulated matrix. Comparing these curves with those in Fig. 2, we see that the Compton edges are positioned at higher pulse heights in the Illinois matrix. This may be caused by inaccuracies in the light units-to-MeVee conversion used for the Illinois matrix.

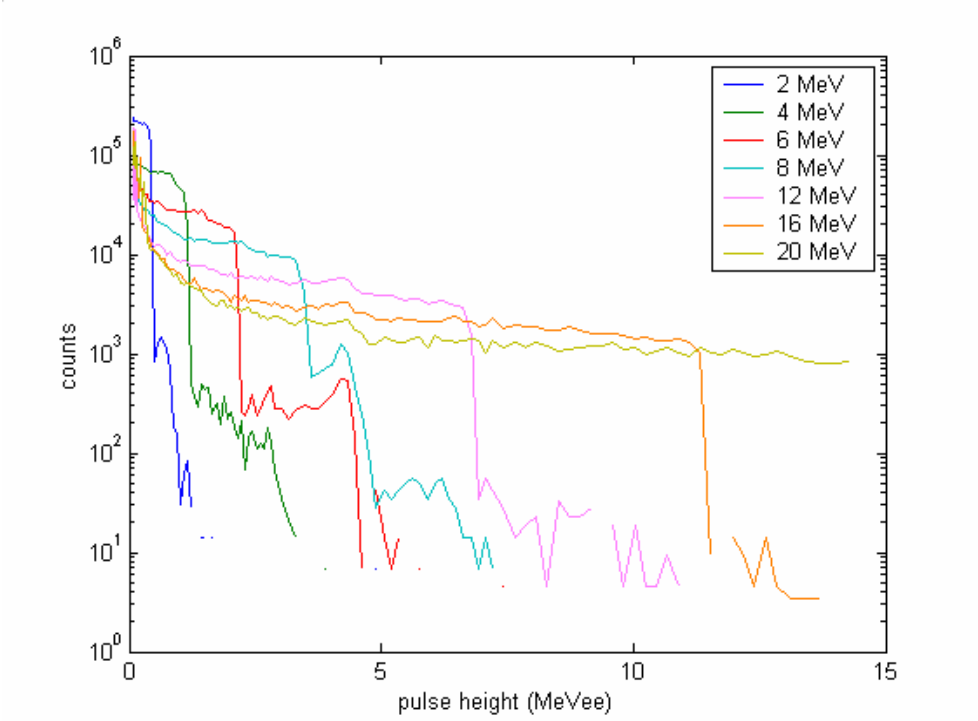

Fig. 4. Some response curves taken from the simulated response matrix. Each column of the matrix corresponds to a different energy.

6. MODIFIED MATRIX

With matrix inversion methods, better results are usually obtained when the response matrix is approximately triangular.[11] For this reason, we eliminated from the matrix the counts above the diagonal defined by the proton recoil edges. In other words, we eliminated from each response function all the counts at pulse heights larger than the proton recoil edge. Those counts are not given by neutrons but by secondary photons produced in the aluminum case of the detector.

Figure 5 shows some response curves taken from the modified matrix.

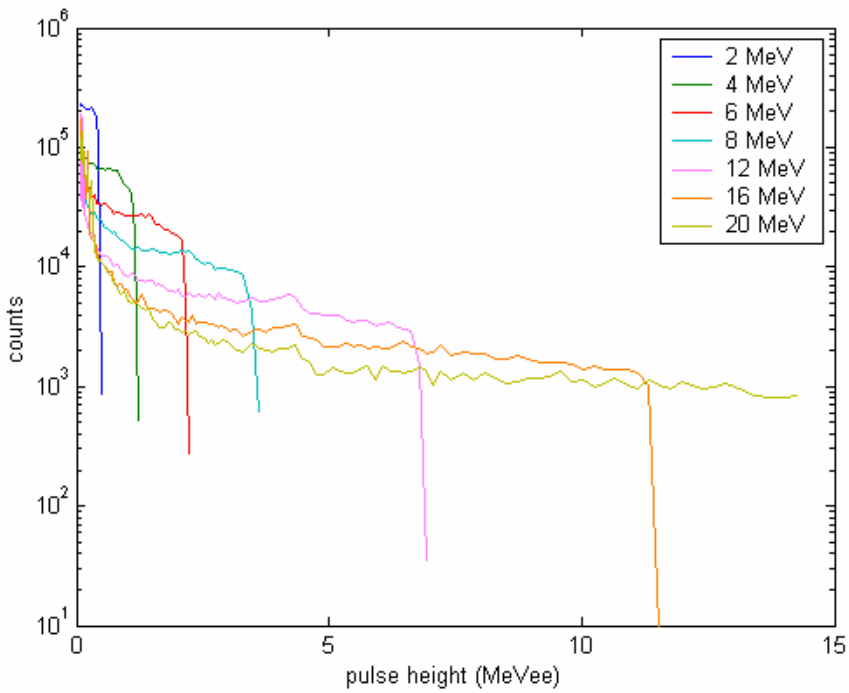

Fig. 5. Some response curves taken from the modified matrix. Each column of the matrix corresponds to a different energy.

7. MEASURED RESPONSE MATRIX

The measurement of the response matrix of the liquid scintillator was performed using a timed Cf-252 spontaneous fission neutron and gamma ray source.[12] The time-of-flight method was used to discriminate neutrons from gamma rays and to determine the neutron energy. In the experiment, the Cf-252 source was placed at a 1-m distance from the liquid scintillator. The data acquisition was performed using a custom-built Matlab program running on a Tektronix TDS-5104 digital oscilloscope. The use of the Matlab Instrument Control Toolbox allowed a totally automated data collection process.

Neutron energies and corresponding pulse heights were stored to build the scintillator's response matrix. The neutron energy was collected in 0.1 MeV bins, within the range 0.5 to 9 MeV (this corresponds to a flight time of 25 to 80 ns, approximately). The pulse heights were collected in 0.05-V bins, in the range 0 to 5.0 V. A total of approximately 100,000 pulses were analyzed.

The measured data were normalized according to the Cf-252 neutron spectrum so that the resulting matrix represents the response of the detector when the number of incident neutrons is the same at each energy. The matrix was also smoothed.

The matrix needs smoothing because statistical errors in the response matrix give large errors and

oscillations in the unfolded spectra.[2] Each response function was smoothed by applying the following method:[11]

$$y_{SMOOTHED}(x) = \frac{1}{4}y(x-1) + \frac{1}{2}y(x) + \frac{1}{4}y(x+1),$$

where x is the bin number and $y(x)$ is the number of counts in bin x.

The total number of counts remains constant with this method, so the normalization of the response functions is conserved. This procedure was repeated twice on each column of the matrix.

Figure 6 shows the measured response matrix for the liquid scintillator.

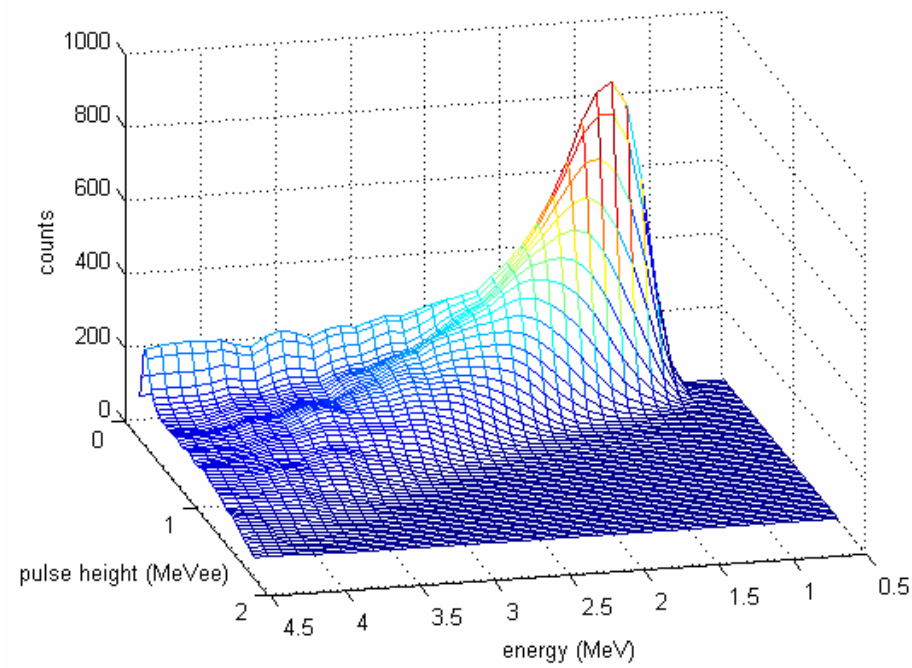

Fig. 6. Measured response matrix.

This measured response matrix is not suitable for use with the FORIST code as it is, because its energy and pulse height limits are too narrow. The Cf-252 source does not emit enough high-energy neutrons.

Figure 7 shows some neutron response curves taken from the measured matrix. A noticeable difference from the simulated response curves in Fig. 6 is that the counts drop almost to zero for very small pulse heights. This happens because only pulses bigger than a threshold were registered. A small number of counts is present in the bins with the lowest pulse height anyway because of the smoothing procedure.

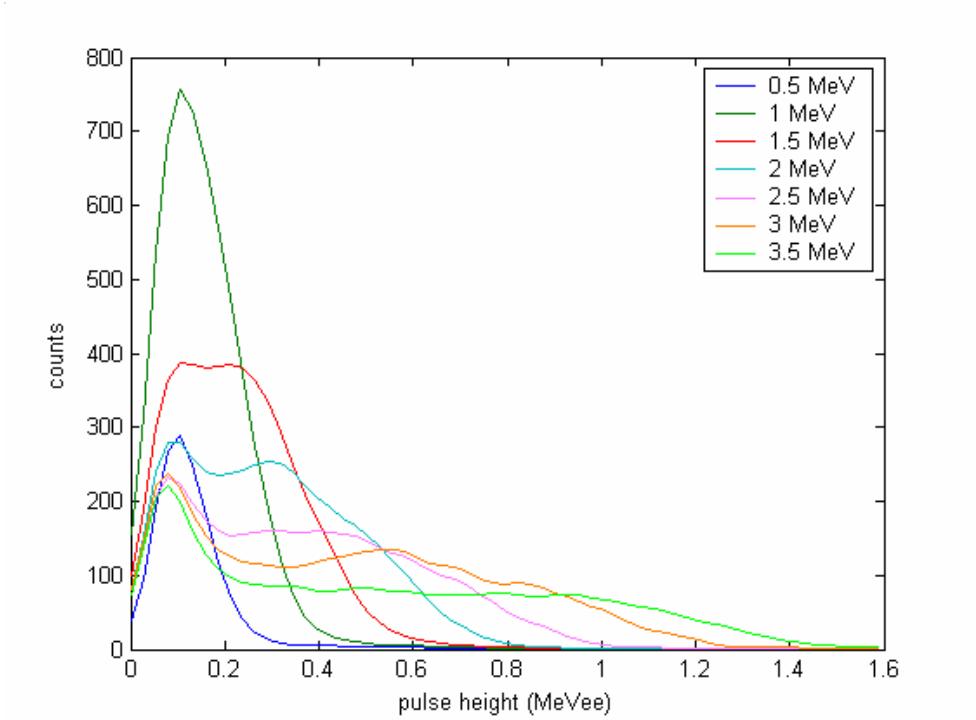

Fig. 7. Some response curves taken from the measured matrix. Each column of the matrix corresponds to a different energy.

8. TEST CASES

In order to test our matrices, we ran several simulations and a measurement, and we unfolded the resulting pulse height distributions.

8.1 Cf-252 NEUTRON SPECTRUM

Our first test case is the simulation of a californium source. For the purpose of testing the neutron unfolding results, we calculated the pulse heights generated in the detector only by incident neutrons (not those generated by the incident photons). Figure 8 shows the neutron spectrum incident on the detector and the spectrum unfolded with the Illinois matrix. The two curves are normalized so that they have the same area between 2 and 8 MeV. There is a reasonable agreement between the two spectra for energies above 3 MeV.

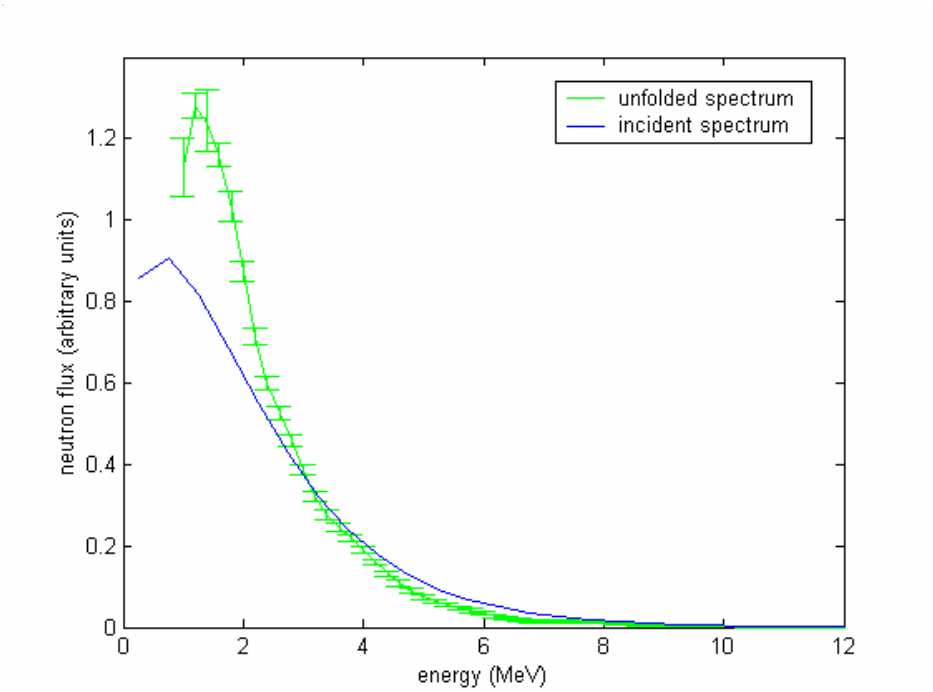

Fig. 8. Cf-252 spectrum unfolded with the Illinois matrix. The error bars indicate plus and minus one standard deviation, as calculated by the FORIST code.

Figure 9 shows the Cf-252 spectrum unfolded with the simulated unsmoothed matrix. The agreement with the incident spectrum is quite good. At low energies (below 1 Mev), the errors are very large, and the solution is not reliable. We see that this unfolded spectrum presents some oscillations, especially at low energies. This is a common problem when spectra are unfolded with matrix inversion methods.[14, 15]

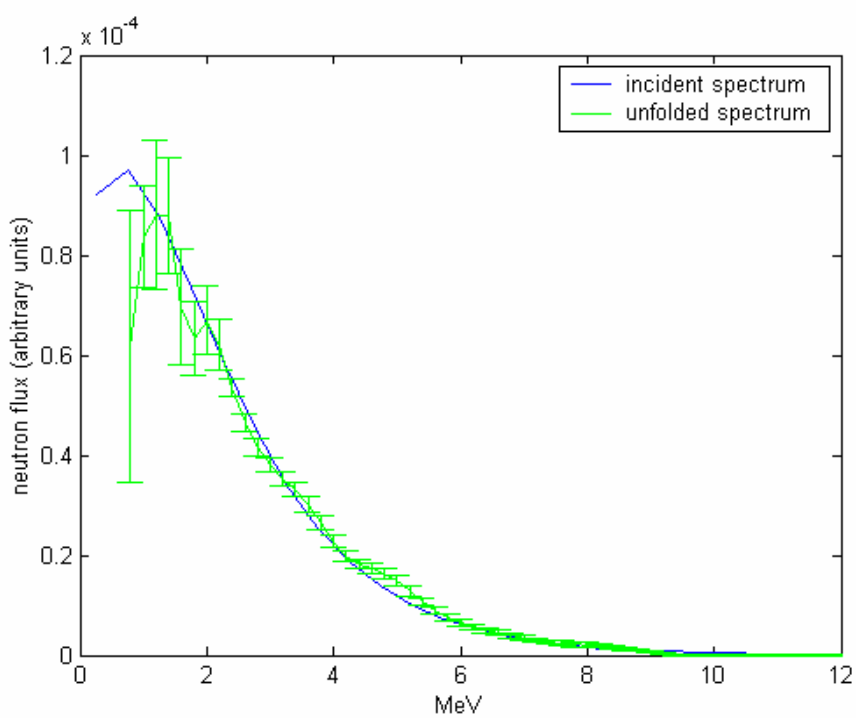

Fig. 9. Cf-252 spectrum unfolded with the simulated matrix.

Figure 10 shows the Cf spectrum unfolded with the modified simulated matrix described in Section 6. If we compare it with Fig. 9, we see that the oscillations at low energies are much reduced and the unfolded spectrum is in good agreement with the incident spectrum.

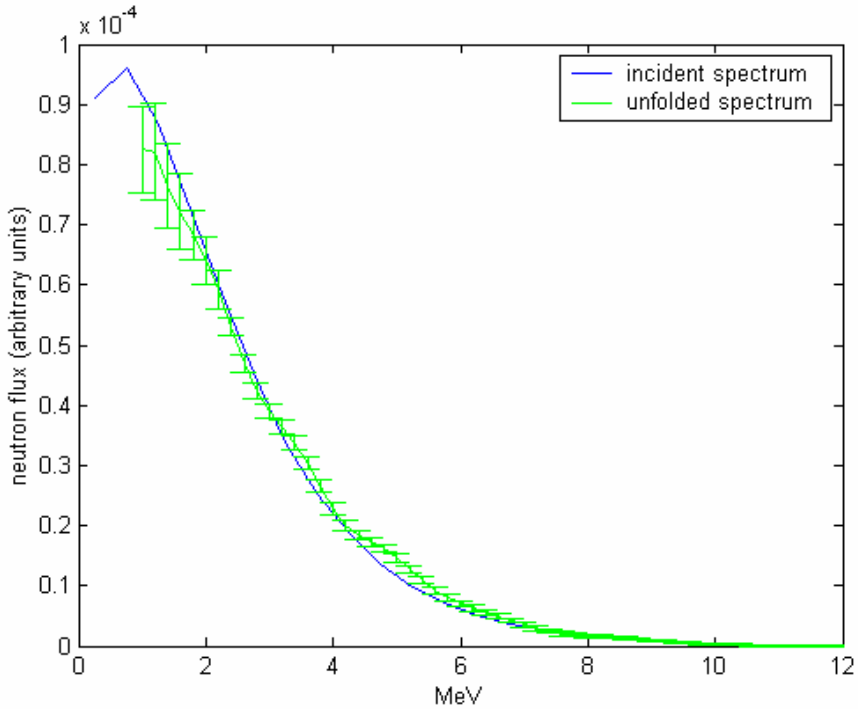

Fig. 10. Cf-252 spectrum unfolded with the modified simulated matrix.

8.2 14-MeV NEUTRONS

Our second test case is a monoenergetic beam of 14-MeV neutrons. Figure 11 shows the result of unfolding with the Illinois matrix. The unfolded spectrum shows a single big peak, as expected, but its energy is 15 MeV instead of 14 MeV. We can also see that the calculated neutron flux is negative at the sides of the peak.

Figure 12 shows the same simulation unfolded with our simulated matrix. There is considerable improvement in the unfolded spectrum: the 14-MeV peak has the right energy, and the calculated flux is positive or zero at all energies. There are some small oscillations at lower energies (below 8 MeV).

Figure 13 shows the simulation unfolded with our modified matrix. The oscillations are almost completely eliminated, and the errors are reduced. The agreement with the incident neutron spectrum is excellent.

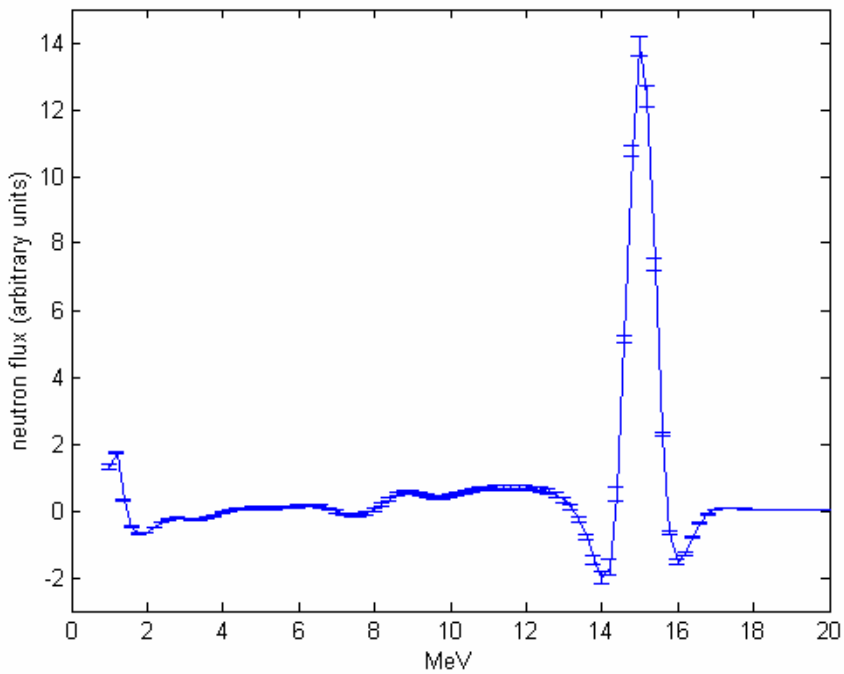

Fig. 11. 14-MeV neutrons, unfolded with the Illinois matrix.

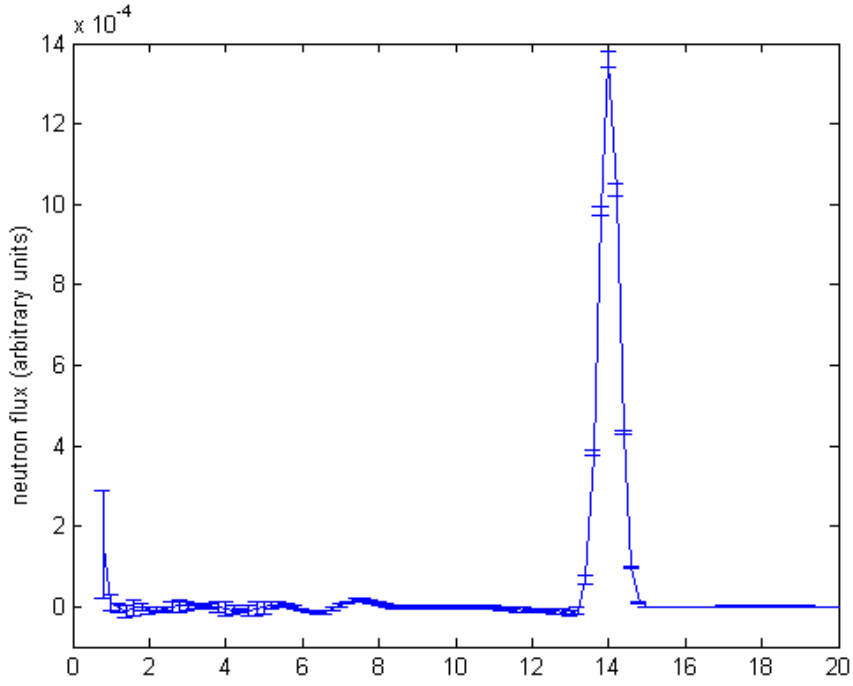

Fig. 12. 14-MeV neutrons, unfolded with the simulated matrix.

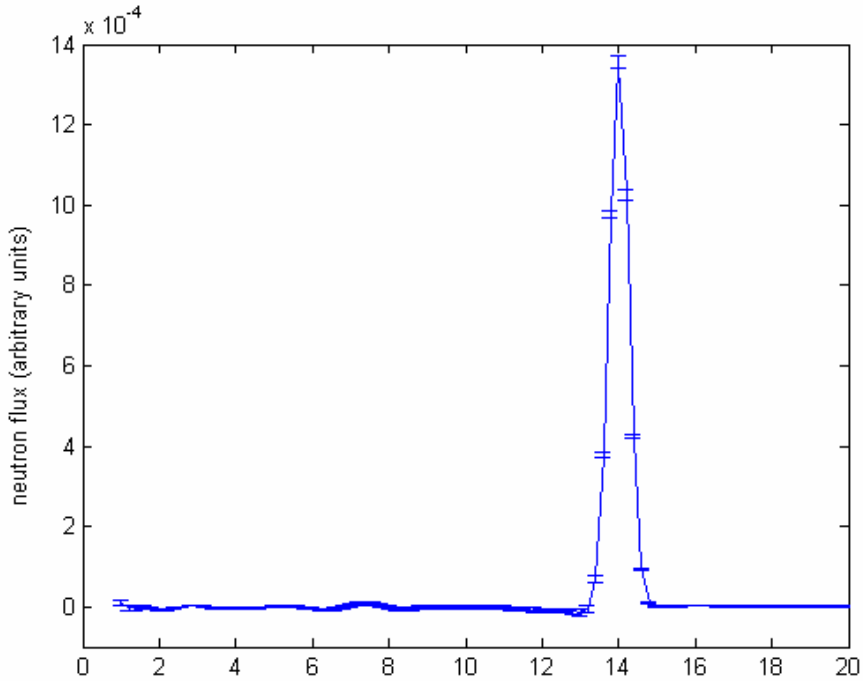

Fig. 13. 14-MeV neutrons, unfolded with the modified simulated matrix.

8.3 4-MeV AND 14-MeV NEUTRONS

Our third test case is the simulation of a neutron source emitting 4-MeV and 14-MeV neutrons.

Figure 14 shows the spectrum unfolded with the Illinois matrix. The two peaks are very well resolved, but their energy is again inaccurate.

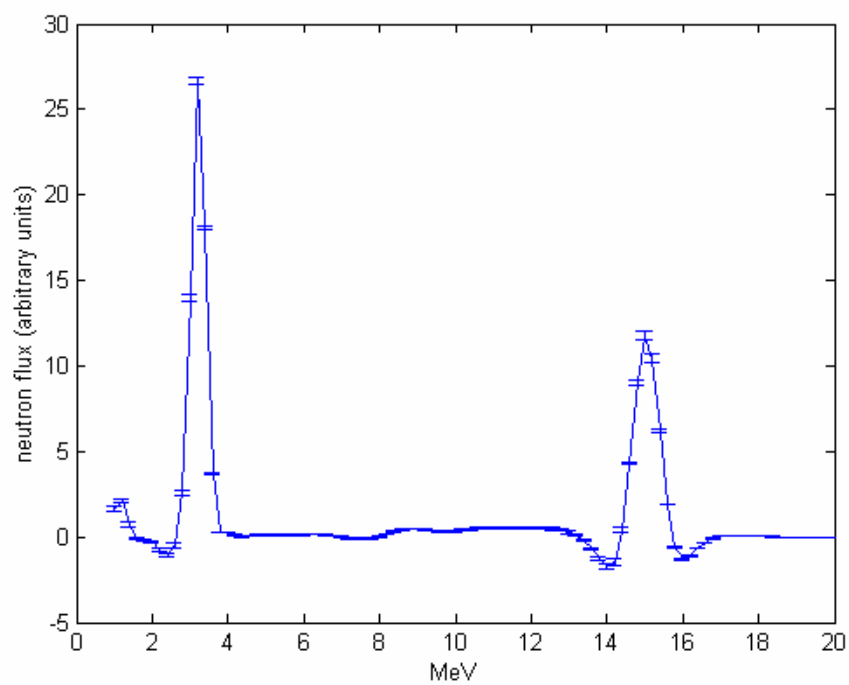

Fig. 14. 4-MeV and 14-MeV neutrons, unfolded with the Illinois matrix.

Figure 15 shows the results of unfolding with our simulated matrix. The 4-MeV peak and the 14-MeV peak have the right energies.

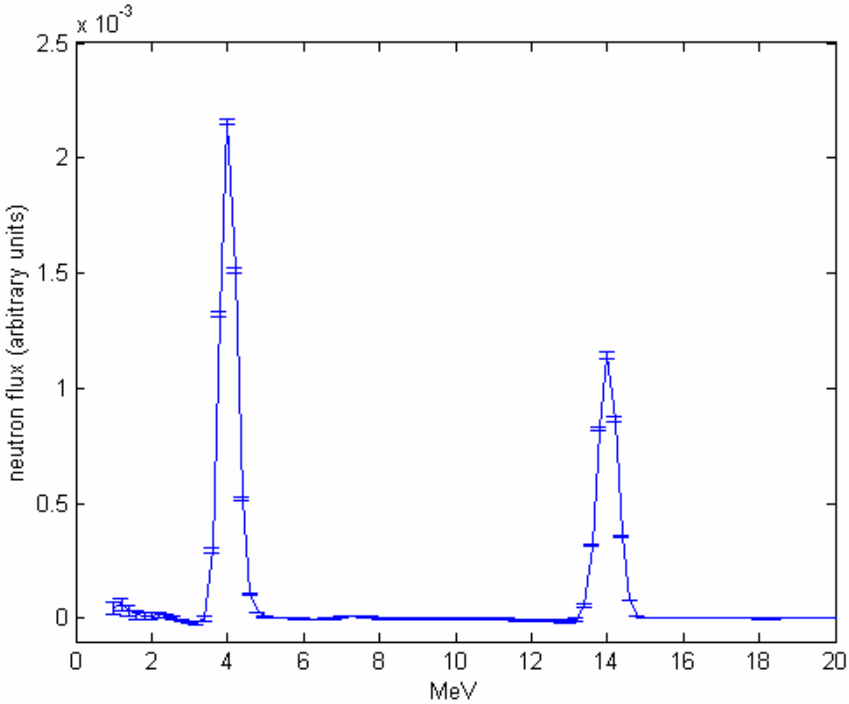

Fig. 15. 4-MeV and 14-MeV neutrons, unfolded with the modified simulated matrix.

8.4 Am-Be NEUTRON SPECTRUM

Our fourth test case is the simulation of an Am-Be source. As for the Cf-source simulation, we calculated the pulse heights generated in the detector by incident neutrons, but not those generated by incident photons.

We unfolded this spectrum with our simulated matrix. Figure 16 shows the unfolded Am-Be neutron spectrum together with the incident neutron spectrum in the simulation. The two curves are normalized so that the area above 1 MeV is the same. The agreement between the two curves is reasonable. Figure 17 shows the Am-Be spectrum unfolded with the modified version of the simulated matrix described in Section 6. The errors are reduced, and the agreement with the incident spectrum is very good.

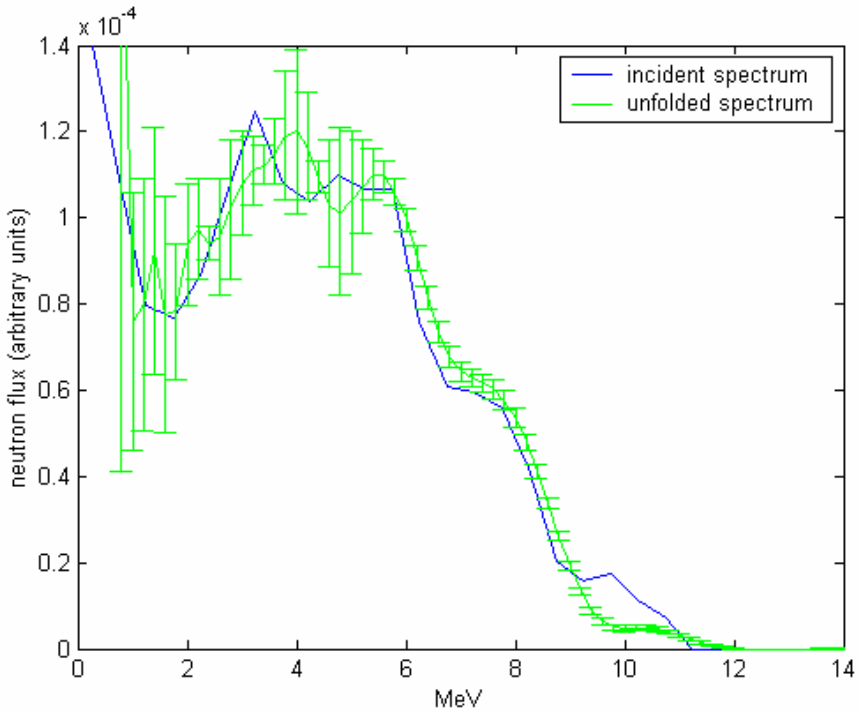

Fig. 16. Am-Be neutron spectrum, unfolded with the simulated matrix.

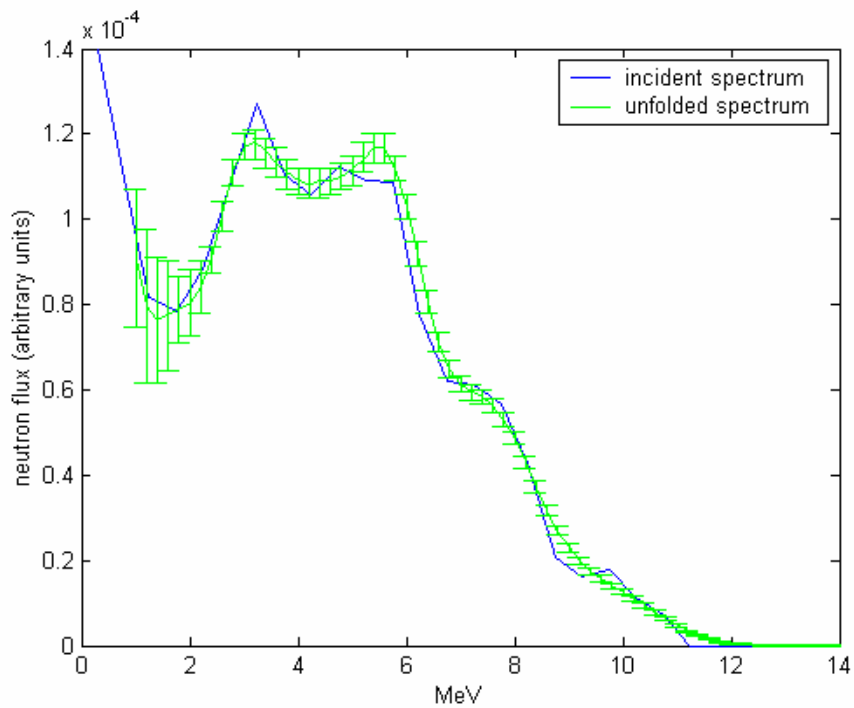

Fig. 17. Am-Be neutron spectrum, unfolded with the modified simulated matrix.

8.5 MEASUREMENT OF Cf-252 SOURCE

Our fifth test case is the measurement of the pulse height spectrum of neutrons from a Cf-252 source. The neutrons were discriminated from the photons by a time-of-flight method. We unfolded this spectrum with our modified simulated matrix, which is the one that gives the best results in the other tests.

Figure 18 shows the unfolded Cf-252 neutron spectrum, together with the incident neutron spectrum. The two curves are normalized so that the area between 1 and 8 MeV is the same. There is reasonable agreement between the two curves for energies greater than 1 MeV.

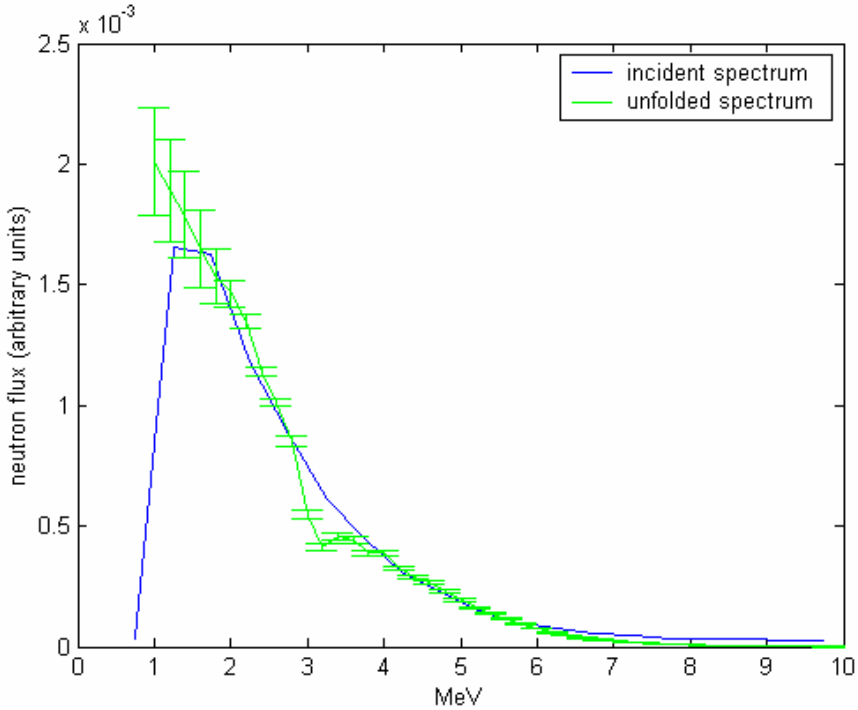

Fig. 18. Measured Cf-252 neutron spectrum.

9. CONCLUSIONS AND FUTURE WORK

We presented some preliminary results of neutron spectrum unfolding using the FORIST code. We obtained good results except in the lower energy part of the spectrum (0-1 MeV). This was also observed by other authors.[15, 16]

We obtained very good results in the unfolding of neutron spectra of monoenergetic neutrons and of radioactive neutron sources, such as Cf-252 and Am-Be. Agreement between the simulated unfolded spectra and the actual spectra was very good with the simulated matrix.

A matrix that is specifically built for the detector used gives better energy accuracy than a response matrix based on other detectors. Even though the scintillating material is the same, the response is affected by the detector's size and the materials around it. Further errors are introduced when the calibration in light units/channel is different for the response matrix and the pulse height spectrum to be unfolded, because it is difficult to calculate the exact conversion factor.

This work shows how the MCNP-PoliMi code could be used to calculate the neutron response matrix of different detector types with great accuracy. This method is faster, more practical, and more economical than directly measuring the detector's response to neutrons of different energies.

The results are encouraging and will be used in conjunction with nuclear safeguards methods to determine the properties of nuclear materials.

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