Localization and spectral isolation of special nuclear material using stochastic image reconstruction

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Abstract

In this work we present a technique for isolating the gamma-ray and neutron energy spectra from multiple radioactive sources localized in an image. Image reconstruction algorithms for radiation scatter cameras typically focus on improving image quality. However, with scatter cameras being developed for non-proliferation applications, there is a need for not only source localization but also source identification. This work outlines a modified stochastic origin ensembles algorithm that provides localized spectra for all pixels in the image. We demonstrated the technique by performing three experiments with a dual-particle imager that measured various gamma-ray and neutron sources simultaneously. We showed that we could isolate the peaks from ^{22}Na and ^{137}Cs and that the energy resolution is maintained in the isolated spectra. To evaluate the spectral isolation of neutrons, a ^{252}Cf source and a PuBe source were measured simultaneously and the reconstruction showed that the isolated PuBe spectrum had a higher average energy and a greater fraction of neutrons at higher energies than the ^{252}Cf . Finally, spectrum isolation was used for an experiment with WGPu, ^{252}Cf , and AmBe. The resulting neutron and gamma-ray spectra showed the expected characteristics that could then be used to identify the sources.

1. Introduction

Radiation imaging is an effective tool for localizing sources of radioactive material by reconstructing source distributions. A specific type of imaging system, radiation scatter cameras, have

long been the subject of study for applications including astronomy, emergency response, and non-proliferation [1]–[7]. Scatter cameras are an attractive tool due to their large field-of-view and ability to characterize source location and energy. However, image reconstruction using simple backprojection produces images with low resolution and low signal-to-noise ratios due to a large range of possible origins for each detected event.

To improve image resolution and signal-to-noise, statistical techniques have been successfully applied to image reconstruction. One of the most widely used methods is list-mode maximum-likelihood expectation-maximization (MLEM), which estimates the source distribution by using a response matrix created from observed events [8]–[11]. List-mode MLEM constructs an image through a number of iterations where the estimate at each iteration increases the value of the likelihood function. While list-mode MLEM provides a large improvement in image quality from simple backprojection, images can suffer from the algorithm over-fitting noise due to incomplete information in the system matrix. A stopping condition is also required as over-iteration is well known to cause degradation in image quality [12].

The stochastic origin ensembles (SOE) method has been proposed by Andreyev et al. [13] as an alternative image reconstruction method to list-mode MLEM that addresses the aforementioned issues. SOE is an iterative method that is based on Markov-chain Monte Carlo through use of the Metropolis-Hastings algorithm [14]. The SOE algorithm was originally applied to tomography and later Compton cameras with literature showing that SOE can produce comparable image quality to list-mode and bin-mode MLEM image reconstructions [13], [15], [16]. In prior work we have also demonstrated the image reconstruction capabilities of SOE for experiments using a dual-particle imaging system to measure ^{252}Cf and mixed-oxide nuclear fuel – which was designated as Category-III special nuclear material (SNM) [16].

Because past application of the SOE algorithm was for medical imaging, radioactive tracers with known emitted energy spectra were used. However, for imaging applications involving SNM, the emitted energy spectra are of great interest as they can be used to identify and discriminate between weapons-usable and non weapons-usable radioactive materials. Aggregate energy spectra for gamma-rays and neutrons are easily reconstructed by the dual-particle imager (DPI) but more advanced analysis is required to isolate the spectra for each source present. In this paper we have modified the SOE algorithm to include a reconstructed energy spectrum for the emitted gamma-rays and neutrons from each pixel in the image. The spectra from pixels corresponding to sources in an image can then be used for source characterization.

Spectral isolation also has the advantage of suppressing environmental background. When the spectrum is isolated from a fraction of the image, in which a source is located, the ratio of source-to-background will be larger. The ratio is improved because in that region, the source strength outweighs the background more than in other regions of the image not containing the source. Including image regions without the source will only add more background, lowering the source-to-background ratio.

This paper presents results from three experiments that demonstrate the ability of the modified SOE algorithm to isolate energy spectra from multiple sources measured simultaneously. An experiment with ^{22}Na and ^{137}Cs was used to measure isolated gamma-ray spectra for two sources. A second experiment using ^{252}Cf and PuBe showed spectrum isolation for a spontaneous fission neutron source and a $^9\text{Be}(\alpha, n)$ neutron source. Finally, an experiment with weapons-grade plutonium (WGPu) metal, ^{252}Cf , and AmBe was performed to show that SOE can discriminate WGPu from the other two sources, which also emit both gamma-ray and neutrons.

2. Methods

In this section of the paper we will briefly describe the DPI, which was the measurement system used to acquire data, and the experimental parameters used in acquisition. We will also describe the

modifications made to the SOE algorithm to reconstruct both images and spectra for each pixel in the image.

2.1 The dual-particle imager

The DPI is a combined Compton and neutron scatter camera. It has a two-plane design in which the front plane is composed of organic liquid scintillators and the back plane is a checkerboard pattern containing liquid scintillators and NaI(Tl) scintillators. The pulse shape discrimination capability of the liquid scintillators, through digital charge integration, allows them to function as scatter plane detectors for both gamma-rays and neutrons [17]. The physics principles and design of the system have been detailed in [5] and [18]. The system is pictured in Figure 1.

In this work, the SOE algorithm uses the reconstructed angle, which defines a cone of possible source locations, and the reconstructed energy for each event recorded by the system. In the context of this work, the terms event and count will be used to refer to a correlated detection in two detectors that can be used to reconstruct both the angle and energy of the incident particle. For gamma-rays, a correlation between a front-plane liquid scintillator and back-plane NaI(Tl) scintillator is required to be recorded as an event. For a neutron to be recorded as an event, a correlation is required between a front-plane liquid scintillator and a back-plane liquid scintillator.

2.2 Spectral isolation using a modified SOE algorithm

The SOE algorithm was used for image reconstruction with the DPI in [16]. However, because the algorithm has been modified in several important ways, a full description of the algorithm is provided in this paper. The steps outlined are performed separately for gamma-rays and neutrons.

- 1) For each event, a single possible source position is sampled on the intersection of the cone, representing possible source locations, and a sphere surrounding the system. This sampled location is referred to as an origin. For two experiments in this work, a long-distance imaging

approximation is used, which locates the apex of each cone at the center of the sphere. In this scenario, the distances from the actual cone apexes to the sphere center are assumed to be sufficiently small compared to the distance that the actual source or sources are located from the system. We define the center of the sphere to correspond to the middle of the gap between both detection planes of the DPI. Because uncertainties exist in the measured quantities, not all cones will overlap with the location of the actual source.

To compensate, the angle for each cone is sampled from a probability density function (PDF) defined as a normal distribution,

$$\mathbf{PDF} = \frac{1}{\sigma\sqrt{2\pi}} e^{\frac{-(x-\theta)^2}{2\sigma^2}} \quad (1)$$

with the calculated angle, θ , as the mean. In a separate analysis, we found that spatial uncertainty, caused by the unknown interaction location within each detector cell, dominated the overall uncertainty. Other factors in the overall uncertainty include the energy and time resolution of detected particles. Sampling each calculated cone angle from a normal distribution with a FWHM of 10° ($\sigma = 4.25^\circ$) for both gamma-ray and neutron events was found to be a sufficient approximation for image reconstruction.

An origin is then sampled on the cone-sphere intersection that is defined by the angle θ , which is sampled from the normal distribution in Equation 1. The sampled origins for each event are then pixelated into a two-dimensional grid. A 5° -pixel size was chosen based on the resolution of the system. The pixelated image created in this step represents the initial image state.

- 2) A histogram is created to record the number of origins that correspond to a particular pixel and energy bin. The total number of separate histogram bins corresponds to the number of pixels multiplied by the number of energy bins. The histograms are populated according to

the numbers of origins contained within a particular pixel and energy bin in the initial image state.

- 3) The origin for each event is resampled using the same process outlined in step one.
- 4) The new location for each origin is then accepted or rejected. The acceptance criterion is based on the number of origins contained within the pixel where the origin was originally located, $\mathbf{p}$, in the prior image state $\mathbf{I}_s$, represented by $\mathbf{N}_{\mathbf{p},s}$, and the number of origins contained within the pixel at the new location of the origin, $\mathbf{p}'$, of the new image state $\mathbf{I}_{s+1}$, represented by $\mathbf{N}_{\mathbf{p}',s+1}$. The acceptance probability for the new location of an origin is defined as:

$$A(\mathbf{I}_s \rightarrow \mathbf{I}_{s+1}) = \min \left(1, \frac{N_{\mathbf{p}',s+1}}{N_{\mathbf{p},s}} \right) \quad (2)$$

Origin locations that have moved to a pixel with a larger number of origins than the pixel for the original location are always accepted. If there are less origins contained within the pixel at the new location the acceptance probability is simply the ratio of the values. This definition of A makes the Markov chain by which image states are formed ergodic.

- 5) Values corresponding to the numbers of origins contained within particular pixel and energy bins are recorded in each histogram.
- 6) Steps three through five are repeated over a number of iterations, with each iteration creating the new image state from the prior image state.
- 7) After a sufficient number of iterations is performed to reach steady-state for all histograms, various solutions can be created from the distributions that estimate the actual source location(s) and spectra. For example, an image for each energy bin can be created by taking the mean or the mode of the distribution for each pixel. These solutions can then be summed to form an image that includes all energies. An energy spectrum for each pixel is formed by using the mode of the distribution for each energy bin. The distributions are fit with a first-order Gaussian distribution using the built-in MATLAB function *fit*. If the fitting fails or

produces a fit with a negative mode, which occasionally occurs for bins with low numbers of origins, the value in the isolated spectrum is set to zero.

Several parameters are required to define the binning structure of the histograms. These parameters include the width of each energy bin, the bin size for recording the number of origins in a histogram for each iteration. A total number of iterations is also required. In this work these parameters were chosen empirically for each experiment to achieve sufficient statistics for isolated spectral reconstruction.

2.3 Experiments

Three experiments were performed with the DPI to show the ability of the described SOE algorithm to isolate energy spectra when multiple sources are present. An experiment using ^{22}Na and ^{137}Cs showed the capabilities of gamma-ray spectrum isolation. An experiment using ^{252}Cf and PuBe was performed to show neutron spectrum isolation. Finally, an experiment with WGPu, ^{252}Cf , and AmBe demonstrated the ability to localize a neutron and gamma-ray SNM spectra in an environment with multiple sources emitting neutron and gamma-rays.

3. Experimental results and discussion

The goal of this work was to demonstrate that image reconstruction using SOE, which offers several advantages over other reconstruction methods, could isolate spectral information from multiple sources measured simultaneously. In doing so, the unique signature of SNM can be identified. The experiments performed were designed to offer insight into spectrum isolation between gamma-ray sources and neutron sources that have different spectral features to show a proof of concept before applying the methodology to a scenario involving multiple radioactive source including SNM.

In this section we present and discuss the experiments used to evaluate spectrum isolation. Relevant gamma-ray and neutron images and isolated spectra reconstructed using the using SOE

algorithm described in section 2.2 are shown. We also present several pixel and energy bin histograms to examine their structure and comment on uncertainty in the isolated spectra.

3.1 ^{22}Na and ^{137}Cs experiment

Two well-known gamma-ray sources were measured with the DPI to evaluate the quality of SOE reconstruction for distinct energy peaks. The setup, results, and a discussion of the experiment follow.

3.1.1 Experimental setup

^{22}Na and ^{137}Cs point sources were measured simultaneously with the DPI for 20 minutes. The ^{22}Na had angular coordinates of (116° azimuthal, 72° inclination) and was located 543 cm from the center of the DPI. The coordinate system was defined such that a source centered directly in front of the DPI has angular coordinates (90° , 90°). The ^{137}Cs was located at (57° , 104°) at a distance of 306 cm from the DPI. The experimental setup is shown in Figure 1. Activities of the sources were 3.27 MBq and 3.24 MBq for the Na^{22} and ^{137}Cs respectively. The ^{22}Na signature consists of two gamma-ray energies at 0.511 MeV and 1.275 MeV. The ^{137}Cs gamma-ray emission of interest is at 0.662 MeV.

Fig. 1. This photo shows the experimental setup for a measurement of ^{22}Na and ^{137}Cs . The sources positions are highlighted by red circles. The ^{22}Na was located at an angular coordinate of $(116^\circ, 72^\circ)$, 543 cm from the DPI and the ^{137}Cs was located at $(57^\circ, 104^\circ)$ at a distance of 306 cm. The DPI is seen at the right of the photo.

3.1.2 Results

In this experiment a total of 54,541 gamma-ray events were detected. We found that using an energy bin width of 10 keV and 1×10^6 iterations provided sufficient statistics for both isolated energy spectra. The histogram for each energy bin/pixel used a bin width of one origin.

The reconstructed image shown in Figure 4 has two clearly defined hot-spots. The ^{22}Na reconstructed to the pixel $(115^\circ, 75^\circ)$ and the ^{137}Cs created a hot-spot occupying the pixels $(55^\circ, 105^\circ)$ and $(60^\circ, 105^\circ)$. Table 1 compares the angular location of the reconstructed hot-spot with the expected angular location of the hot-spot. To identify the expected location for the reconstructed hot-spot, the actual angular locations of the sources were rounded to bin centers of the nearest pixel. The image was created using the mean value for each pixel over all image states. Because the ^{137}Cs was located near the edge of a pixel, contribution is seen in two pixels and makes the intensity appear lower than if the contribution were located in a single pixel such as for the ^{22}Na .

Table 1. The expected angular locations of the ^{22}Na and ^{137}Cs are compared to the locations of the most intense pixel in the reconstructed hot-spots. The angular distance from the expected source location to the reconstructed source location is given.

	^{22}Na		^{137}Cs	
	Azimuth	Inclination	Azimuth	Inclination
Expected location	115°	70°	55°	105°
Reconstructed location	115°	75°	55°	105°
Absolute difference	0°	5°	0°	0°

The isolated spectra shown in Figure 5 were created from a 3×3 -pixel region surrounding the most intense pixel in the hot-spots. The nine pixel regions ensured that the full signal was captured when there was contribution in multiple pixels. For comparison, the reconstructed spectrum from all recorded events is shown on the same plot. In the total spectrum, the resolution of the 0.511-MeV, 0.662-MeV, and 1.275-MeV peaks were 10.6%, 9.4%, and 12.0% respectively. These resolutions were lower than the expected 15% for the 0.511-MeV peak and 13% for the 0.622-MeV peak. The resolution was greater than the expected 9% for the 1.275-MeV peak. A discussion of the discrepancy follows in Section 3.1.3.

Fig. 4. The reconstructed gamma-ray image shows two hot-spots which locate the ^{22}Na and ^{137}Cs sources. The ^{22}Na is located by the more intense pixel at top left. The ^{137}Cs hot-spot, in the lower right, appears less intense because it is split between two adjacent pixels.

Fig. 5. Two isolated spectra from the ^{22}Na and ^{137}Cs hot-spots are shown with the total measured spectrum. This figure shows that the total spectrum is the combination of a ^{22}Na and ^{137}Cs spectrum. The isolated spectra are plotted against the left axis while the total spectrum was plotted against the right axis. Each isolated spectrum was created from the 3×3 -pixel region surrounding the brightest pixel in each hot-spot. The uncertainty is shown by the light shaded regions for the ^{22}Na and ^{137}Cs spectra although it is difficult to see because in most places it is not thicker than the line.

For the 3×3 ^{22}Na region (centered on pixel $(115^\circ, 75^\circ)$), the histogram corresponding to the energy bin 0.51 MeV is shown in Figure 6(a). The mode of the fitted distribution is 105.5 origins, which is then used as the number of counts in the 0.51-MeV energy bin for the spectrum. For the 1.27-MeV energy bin, the mode of the fit, shown in Figure 6(b), was 30.5 origins. The uncertainty for each bin is given by the standard error of the fitted distribution. The resolution of the 0.511-MeV peak in the isolated spectrum was 11.1% and was 7.9% for the 1.275-MeV peak. Section 3.1.3 explains why the reconstructed images were created using mean values and the isolated spectra were created using the mode of the fitted histograms.

Fig. 6. Histograms from the 3×3 -pixel ^{22}Na region for the 0.51-MeV (a) and 1.27-MeV (b) bins are fit well by a first order Gaussian. The histograms represent the number of occurrences for a specific number of origins from the 3×3 region from all 1×10^6 image states. The mode of the fit for the histogram from each energy bin is used to create the isolate spectrum with the standard deviation in each histogram representing the uncertainty.

The histograms corresponding to the 0.66-MeV energy bin of the ^{137}Cs region is shown in Figure 7. For the 0.66-MeV energy bin the mode of the fit was 159.7 origins. The resolution of the isolated spectrum at 0.662 MeV, 10.1%, was similar to that of the peak in the aggregate spectrum.

Fig. 7. The histogram from the 3×3 -pixel region surrounding the ^{137}Cs hot-spot for the 0.66-MeV energy bin was fit with a first order Gaussian. The mode of the fit was used for the 0.66-MeV bin in the isolated spectrum with the standard deviation of the fit representing the uncertainty.

The isolated ^{22}Na spectrum shows the two expected peaks at 0.511 MeV and 1.275 MeV and the isolated ^{137}Cs spectrum shows the expected peak at 0.662 MeV. However, there is some contamination in each spectrum from the other spectrum. For example, a small peak can be seen in the ^{22}Na spectrum at 0.662 MeV and a small peak can be seen in the ^{137}Cs spectrum at 0.511 MeV. Contamination from the 1.275-MeV ^{22}Na peak is not seen in the isolated ^{137}Cs spectrum.

3.1.3 Discussion

In this experiment we evaluated spectrum isolation for two sources emitting distinct gamma-ray decays energies. For this experiment and the following ones, the reconstructed images were created by using the mean value of the distributions while the values in the isolated spectra were populated using the mode of the distribution. For many bins, the mean and mode are the same because the distribution is well fit by a Gaussian. However, in distributions with a lower mean and mode, the data does not form the entire Gaussian distribution. In these cases, the mode of the data will be a smaller value than the mean. We found that using the mean values for image reconstruction produced less artifacts and noise than the mode values in all three experiments. A possible cause of the noise are poor fits for bins with small value modes. Peaks in the isolated spectra should be unaffected by poor fitting at low values because the peaks have large mode values.

The most intense peaks in ^{22}Na and ^{137}Cs were located at 0.511 MeV and 0.662 MeV respectively. The aggregate reconstructed spectrum from the measurement (Figure 5) showed that the DPI had sufficient resolution to resolve the two peaks. The measured resolution values were lower than expected based on the resolution functions for liquid and NaI(Tl) scintillators [19], [20]. Uncertainty in the resolution of the liquid scintillators at low energies was likely the cause of the discrepancy. In [19], the lowest energy measured in the resolution function was about 250 keVee. The resolution

function was extrapolated to lower values. Many of the measured events in the system had energy depositions in the liquid scintillators below 100 keVee. At these low energies, the extrapolated resolution function likely has a large uncertainty.

We also found that the measured resolution in the aggregate spectrum was maintained in the isolated spectra. The resolution degraded in the isolated spectra for the 0.51-MeV and 0.662-MeV peaks by 0.5% and 0.6% respectively. We do not believe these deviations to be significant but a more rigorous investigation including additional gamma-ray decay energies with higher statistics is warranted by the results from this experiment. For the 1.275-MeV peak the resolution was 12.0% in the aggregate spectrum but improved to 7.9% in the isolated spectrum. This difference is probably due to uncertainty in the calculated resolution in the aggregate spectrum. The peak was not much larger than the continuum. The resolution functions for the liquid and NaI(Tl) scintillators stipulate that resolution should be improved at higher energies. The 7.9% resolution calculated in the isolated spectrum is more in line with the expected 9% resolution.

The source localization was also good, with the most intense pixel in the ^{137}Cs hot-spot reconstructing to the expected pixel. The most intense pixel in the ^{22}Na hot-spot reconstructed to the pixel directly below the expectation with a small contribution in the expected pixel. Because the simple backprojection image also has the most intense pixel in the ^{22}Na hot-spot in the same location, this suggests that the shift is attributable to other factors in the measurement, such as the uncertainty in the source positioning, and not the SOE algorithm.

While the energy resolution may suffer a small amount of degradation when isolated, the spatial resolution of the system was the cause for contamination of one spectrum into another. Small but statistically significant peaks are seen at 0.511 MeV in the ^{137}Cs spectrum and 0.662 MeV in the ^{22}Na spectrum (Figure 5). The contamination of events from the other source appearing in each isolated spectrum is because some of the cones defining possible source locations overlapped both actual

source locations. The overlap allowed for a single event to be sampled at either location and is attributable to the broadening of the cone, which represents the uncertainty of the events caused by system resolution. For a theoretical system with improved resolution, the cones would be less broad thereby lowering the probability of overlap between both sources locations.

The proximity of both sources will also factor into the amount of contamination. A small distance between the sources will increase the probability of cones overlapping both events. Quantification of the contamination in an isolated spectrum due to source proximity and system resolution is a topic that will be further studied in the future.

Another observation made in the isolated spectra was the absence of low-energy events that dominate the aggregate spectrum. The isolated spectrum only examines events from a region of interest, which suppresses events from the background radiation that is spread more evenly throughout the environment. This observation highlights one of the main advantages of radiation imaging compared to detection without localization, which is a greater signal-to-noise ratio in the region that the source is located compared to the entire environment.

3.2 PuBe and ^{252}Cf experiment

While many radioisotopes that emit gamma-rays do so with distinct energy-peaks, neutron sources typically emit neutrons with a continuous energy spectrum. Two such sources, PuBe and ^{252}Cf were measured to evaluate performance of the algorithm.

3.2.1 Experimental setup

PuBe and ^{252}Cf point sources were measured simultaneously because they emit different neutron energy spectra. PuBe produces neutrons through the $^9\text{Be}(\alpha, n)$ reaction. The emitted energy spectrum has a different shape and higher average energy than the neutrons emitted by spontaneous fission from ^{252}Cf . The PuBe was located at the angular coordinates $(136^\circ, 92^\circ)$ at a distance of 344 cm from the DPI. The ^{252}Cf was placed at $(71^\circ, 90^\circ)$ with a distance of 419 cm from the DPI. Figure 2 shows

the setup of the measurement. The PuBe had an activity of about 37 GBq and the ^{252}Cf had an activity of about 165 MBq. These activities corresponded to an approximate emission of 1.5×10^6 neutrons per second from the PuBe and 1.9×10^7 neutrons per second from the ^{252}Cf . Because the ^{252}Cf emitted many more neutrons per second than the PuBe, it was shielded with 23.5 cm of polyethylene to avoid masking the PuBe. The measurement time for this experiment was 45 minutes.

Fig. 2. The experimental setup for the measurement of a ^{252}Cf and PuBe source is shown in this photo. Both sources are highlighted by red circles. The ^{252}Cf was located at an angular coordinate of $(71^\circ, 90^\circ)$ and distance of 419 cm from the DPI. A total thickness of 23.5 cm of polyethylene was used to shield the ^{252}Cf so the much greater neutron emission rate would not mask the PuBe source. The PuBe was located at $(136^\circ, 92^\circ)$, 344 cm from the DPI. The DPI appears on the left side of the photo.

3.2.2 Results

In our experiment to demonstrate neutron spectrum isolation, we detected a total of 17,298 neutron events from the simultaneous measurement of PuBe and ^{252}Cf . Figure 8 shows the reconstructed neutron image as an average of all SOE image states. In this image, we see a distinct

hot-spot for the PuBe in pixel (130°, 90°). However, because of the large amount of polyethylene shielding used for the ^{252}Cf , the hot-spot is not as intense and occupies a much larger region. The maximum image value of the hot-spot is at the (80°, 90°) pixel. Table 2 shows a comparison between the expected hot-spot locations and reconstructed hot-spot locations. This image was created using 1×10^6 iterations and a 0.1-MeV energy bin width.

Fig. 8. The reconstructed neutron image shows an intense hot-spot for the PuBe and a less intense, blurred hot-spot for the shielded ^{252}Cf . The ^{252}Cf hot-spot is blurred because 23.5 cm of polyethylene was used as shielding.

Table 2. The expected angular locations of the PuBe and ^{252}Cf sources are compared to the locations of the most intense pixel in the reconstructed hot-spot. The absolute difference between the two locations is given.

	PuBe		^{252}Cf	
	Azimuth	Inclination	Azimuth	Inclination
Expected location	135°	90°	70°	90°
Reconstructed location	130°	90°	75°	90°
Absolute difference	5°	0°	5°	0°

The isolated spectra for each source is shown in Figure 9. The spectrum for the PuBe was created from the 3×3-pixel region surrounding (130°, 90°). The spectrum for the ^{252}Cf was from the 3×3 region centered at (80°, 90°). The histograms shown in Figure 10 correspond to the energy bin producing the maximum value for each region. For PuBe this bin was 2.6 MeV and for ^{252}Cf this bin was 2.4 MeV. Each isolated spectrum was normalized to an integral value of one for comparison, which is shown in Figure 11. The isolated PuBe spectrum had a larger portion at higher energies compared to the isolated ^{252}Cf spectrum as expected. The average energy of the isolated PuBe spectrum was 3.85 MeV and was 2.90 MeV for the isolated ^{252}Cf spectrum.

Fig. 9. The isolated spectra for the PuBe and ^{252}Cf regions are shown in comparison to the total measured spectrum.

The light shaded regions for the isolated spectra represent one-sigma uncertainty calculated from the solution histograms.

Fig. 10. A first order Gaussian was fit to the solution histograms for each energy bin in each 3×3-pixel region. The histograms for the energy bins 2.6 MeV (a) and 2.4 MeV (b) are shown for the PuBe and ^{252}Cf regions respectively. The mode and standard deviation of the fit for each energy bin was used to create both isolated spectra.

Fig. 11. A comparison between the integral-normalized isolated spectra highlights the differences in shape. The ^{252}Cf spectrum has a greater fraction of neutrons at lower energies than the PuBe spectrum. A PuBe source emits a larger fraction of neutrons at greater energies than a ^{252}Cf source so this result was expected. The average reconstructed energy of the ^{252}Cf spectrum was 2.90 MeV and the average energy of the PuBe spectrum was 3.85 MeV.

3.2.3 Discussion

The experiment with PuBe and ^{252}Cf showed the performance of spectrum isolation for two sources with different neutron spectra. There are two key differences in the analysis of this experiment compared to the prior ^{22}Na and ^{137}Cs experiment. First, the neutron spectra emitted by both sources are continuous unlike the gamma-ray spectra from ^{22}Na and ^{137}Cs . Second, there is very little environmental background present in the neutron signal.

The average energy calculated from the isolated ^{252}Cf spectrum was 2.90 MeV, which was higher than the 2.1-MeV average neutron energy emitted from ^{252}Cf . The spectrum in this experiment was also softened compared to the isolated ^{252}Cf spectrum in the WGPu, ^{252}Cf and AmBe experiment. The softened spectrum was caused by neutrons that had been moderated in the large amount of polyethylene shielding the source. Although the reconstructed energy spectrum was softened, the average energy calculated from the isolated spectrum is still larger than the expected average energy of emitted neutrons. A detection threshold for correlated events causes the overall hardening of the reconstructed spectra compared to the emitted spectra. The minimum threshold for a detection in a liquid scintillator was set at 0.04 MeVee which corresponds to a neutron energy deposition of approximately 0.38 MeV. This threshold is required to achieve acceptable pulse shape discrimination between the neutron and gamma-rays. Because a double scatter is required for detection, the minimum energy that can be detected by the DPI is 0.76 MeV. However, the probability of neutrons near this energy scattering such that the energy deposited is above threshold in both detectors is low. Methods to unfold these effects, which would produce a spectrum more representative of the emitted spectrum are under investigation.

Higher energy regions of the isolated spectra are also suppressed due to energy dependent efficiencies. This is especially apparent in the isolated PuBe spectrum which falls off at higher energies much quicker than the emitted spectrum. The DPI is most efficient at about 2.5 MeV after which neutrons of a higher energy are less likely to interact in the detectors. An upper limit also exists due

to the 2-V dynamic range of the digitizer. For this experiment the upper threshold was 3.1 MeVee and for the $^{22}\text{Na}/^{137}\text{Cs}$ and the WGPu/ ^{252}Cf /AmBe experiments the upper thresholds were 2.8 MeVee and 3.1 MeVee respectively. The different upper threshold in the $^{22}\text{Na}/^{137}\text{Cs}$ measurement was due a higher baseline used in waveform acquisition. Again, unfolding techniques may be able to remove this effect from the spectrum to better match the emitted spectrum.

Both sources were localized to the pixel adjacent to the expected pixel. For the PuBe, the most intense pixel in the SOE image hot-spot was in the same location in the simple backprojection image suggesting a bias or uncertainty in the measurement itself and not SOE. However, this is not the case for the ^{252}Cf hotspot although there is still a large contribution in the backprojection image in the same pixel where the SOE image located the source. The difference in location in this case may be due to the broadened hot-spot caused by the large amount of shielding used.

3.3 WGPu, ^{252}Cf , and AmBe experiment

The two prior experiments showed that spectral isolation was effective for gamma-ray energy peaks and continuous neutron fission or (α ,n) spectra. In this experiment both gamma-ray and neutron spectral isolation are used to isolate the spectra from Category-I SNM. We discuss some of the features in the isolated spectra and how they differ from the other two non-SNM sources.

3.3.1 Experimental Setup

To demonstrate gamma-ray and neutron spectrum isolation for Category-I SNM, we used the DPI for experiments at the Device Assembly Facility (DAF) located within the Nevada National Security Site. The DAF provided a 4.1-kg disk of WGPu that measured 10.64 cm in diameter and had a thickness of 2.52 cm. We measured the WGPu in the presence of two other neutron and gamma-ray emitting sources. The WGPu had the coordinates (90° , 85°) and had an emission of approximately 5.2×10^5 neutrons per second. The ^{252}Cf was located at (90° , 109°) and had an emission rate of approximately 3×10^5 neutrons per second. The AmBe was placed at (141° , 85°) and emitted

approximately 1×10^6 neutrons per second. The WGPu, ^{252}Cf , and AmBe sources were located 201 cm, 212 cm, and 207 cm from the DPI respectively and were measured for 850 minutes. Figure 3 shows the sources in relation to the DPI. A 1.3 cm lead shadow-shield was placed 30 cm in front of the WGPu to help reduce the large flux of low energy gamma-rays – particularly the 0.0595-MeV emission from built up ^{241}Am , which has a specific activity of 4.54×10^{10} gamma-rays per second per gram. The AmBe was also shielded with about 10 cm of lead to accommodate another detection system that was measuring concurrently with the DPI.

Fig. 3. The three sources are highlighted by red circles in these pictures of the experimental setup. The WGPu had the angular coordinate (90°, 85°), the ^{252}Cf (90°, 109°), and the AmBe (141°, 87°). The three sources were located at distances of 201 cm, 212 cm, and 207 cm from the DPI respectively.

3.3.2 Results

The reconstructed neutron image shows three hot-spots in Figure 12. To generate this reconstruction, 1×10^5 iterations were used with 0.10-MeV energy bin widths and a total of 161,189 neutron events. Due to the closer proximity of the sources to the DPI, the long-distance approximation was not used for reconstruction in this experiment. Each cone was projected onto a

sphere with a radius of 200 cm. The expected hot-spot locations are compared to their reconstructed locations in Table 3.

Fig. 12. The reconstructed neutron image shows three hot-spots, localizing the WGPu, ^{252}Cf , and AmBe. The left-most hot-spot is the AmBe source, the upper-right most is the WGPu, and the lower-right most is the ^{252}Cf .

Table 3. A comparison between the expected angular location for each source and the actual angular location is given. The absolute difference between the two locations is also shown.

	WGPu		^{252}Cf		AmBe	
	Azimuth	Inclination	Azimuth	Inclination	Azimuth	Inclination
Expected Location	90°	85°	90°	110°	140°	85°
Reconstructed neutron location	90°	85°	90°	105°	135°	85°
Reconstructed gamma-ray location	90°	85°	90°	110°	N/A	N/A
Absolute difference (neutrons)	0°	0°	0°	5°	5°	0°
Absolute difference (gamma-rays)	0°	5°	0°	0°	N/A	N/A

The isolated neutron spectra, shown in Figure 13(a), for each hot-spot for the WGPu, ^{252}Cf , and AmBe were generated from 3×3-pixel regions centered at (90°, 85°), (90°, 105°), and (135°, 85°) respectively. The spectra were normalized for comparison and are shown in Figure 13(b). The shapes

for the WGPu and ^{252}Cf hot-spots are similar as expected, with average energies of 3.23 MeV and 3.20 MeV respectively, since both sources emit neutrons in a Watt distribution. However, the AmBe spectrum has a different shape with a larger fraction of neutrons being emitted at higher energies and an average energy of 3.85 MeV.

Fig. 13. The isolated neutron spectra for the 3×3 -pixel regions surrounding each hot-spot are compared using absolute number of origins (a) and normalized number of origins (b) with the light shaded regions showing one-sigma uncertainty. The spectra for the WGPu and ^{252}Cf are similar because both sources emit neutrons in a Watt distribution. The average energies of the WGPu and ^{252}Cf spectra were 3.23 MeV and 3.20 MeV respectively. The average energy of the AmBe spectrum was greater as expected at 3.85 MeV.

The reconstructed gamma-ray image, in Figure 14, showed only two hot-spots, which located the WGPu and the ^{252}Cf . A hot-spot is not seen for the AmBe because of the 10-cm lead shield placed in between it and the DPI. A total of 1×10^5 iterations was used in the reconstruction.

Fig. 14. The reconstructed gamma-ray image shows two hot-spots for the WGPu (upper) and ^{252}Cf (lower). A hot-spot for the AmBe is not visible because 10 cm of lead was used to shield the source.

The isolated spectra were taken from 3×3 -pixel regions centered at $(90^\circ, 85^\circ)$ and $(90^\circ, 110^\circ)$ and are shown in Figure 15(a). Table 3 compares the actual source locations with the reconstructed hot-spot locations. A comparison of the normalized spectra is shown in Figure 15(b). The spectrum isolated from the WGPu showed a peak at 0.64 MeV that is less prominent in the isolated spectrum from ^{252}Cf . The peak in the WGPu spectrum is a result of decay gamma-rays from ^{239}Pu and ^{241}Am , which were both isotopes present in the sample. The WGPu contained approximately 205 g of ^{240}Pu which has a specific activity of 1.05×10^3 for the 0.642 keV gamma-ray. The peak at 0.62 MeV in the ^{252}Cf spectrum was due to fission products that built up in the source and some contamination from origins that were actually produced from the WGPu. The ^{252}Cf had an approximate age of 10 years. A feature was also seen in the ^{252}Cf spectrum at about 0.48 MeV. This feature can be attributed to photon production from the (n, α) reaction on ^{10}B , which composed part of the optical windows in the EJ-309 liquid scintillators. This feature is not seen in the WGPu spectrum because the source-to-background ratio is larger so it is washed out and other gamma ray energies are present in this region.

Fig. 15. A comparison between the isolated spectra from the 3×3-pixel regions for WGPu and ²⁵²Cf is shown in absolute (a) and normalized (b) scales. The WGPu spectrum display two prominent peaks at 0.38 MeV and 0.64 MeV, which are characteristic of Pu and Am gamma-ray decay energies. The ²⁵²Cf spectrum also has two peaks at these energies, likely due to the build-up of fissions products, but with much lower intensities than the peaks in the WGPu spectrum. The shoulder at 0.48 MeV in the ²⁵²Cf spectrum is from the (n,α) reaction on ¹⁰B. This feature is not seen in the WGPu spectrum because of other gamma-rays decay energies present near this energy and the source-to-background ratio is larger.

3.3.3 Discussion

In this experiment we were able to demonstrate that the SOE algorithm could be used as a first step towards discriminating and identifying SNM in an environment where other radioactive sources are present. In this case, it was significant for us to show that the neutron spectra for WGPu and ²⁵²Cf had the same shape (Figure 13(b)) and average energy. Of course further steps would be required to make a robust identification – especially if shielding has changed the spectral shape. Unfolding the isolated spectra would be the another step in identification because it would remove the resolution and efficiency effects found in the reconstructed spectra. This would transform the isolated spectra into a closer representation of the actual emitted spectra.

This experiment also showed that a source completely invisible to gamma-ray detection by the DPI was detected and imaged using the neutron signal, which demonstrates the advantage of dual-

particle sensitivity. A system detecting both particle types has a better chance of finding all present sources of SNM in an environment, especially if shielding is present.

Using the gamma-ray signal also allowed us to identify a difference in the signal produced by the WGPu and the ^{252}Cf . The normalized gamma-ray spectra (Figure 15) show features that can be used to identify and discriminate WGPu from other gamma-ray sources. Two large peaks are seen in the WGPu at 0.38 MeV and 0.640 MeV which are not prominent in the ^{252}Cf spectrum. There are two notable features seen in the ^{252}Cf spectrum that may make identification more difficult. The first is a shoulder at about 0.48 MeV from gamma-rays created by the (n,α) reaction on ^{10}B , present in the liquid scintillator optical windows, and the second is a small peak at 0.62 MeV from built-up fission products and a contamination from WGPu origins. Due to the age of the source, ^{249}Cf is also present in the sample which emits a 0.388-MeV gamma-ray with an intensity of 66%. Further characterization of the SOE algorithm may provide a method for quantification and subtraction of contamination of origins in the wrong spectrum. We must also seek to understand the effect of gamma-ray production from neutron interactions in our detectors when a large field of slower neutrons is present.

In this experiment, the source localization was consistent with the prior two experiments. In future work we will seek to combine the neutron and gamma-ray cones to generate a combined SOE reconstruction. This may assist with some of the localization discrepancies seen in these experiments. Using smaller pixels may also provide more insight into the accuracy of the localization, however, finer pixels require more iterations for convergence making the problem more computationally expensive.

4. Conclusions and Future Work

The experiments detailed in this work demonstrate the powerful combination of SOE spectrum isolation for image reconstruction with the DPI. For non-proliferation applications, completeness (finding all sources) and characterization (identifying sources of interest) are essential. Our modified

SOE algorithm is able to successfully isolate spectra from multiple hot-spots in an image, which allows for further analysis to identify sources. We demonstrated this method in several experiments that included sources emitting both gamma-rays and neutrons, including an experiment in which we isolated the spectra from WGPu from two other sources.

In our experiment measuring two gamma-ray sources, we showed that we could resolve the 0.511-MeV and 0.662-MeV peaks from ^{22}Na and ^{137}Cs achieving 11.1% and 10.1% resolutions in the isolated spectra respectively. For neutron spectrum isolation, we calculated the average energy of the isolated ^{252}Cf spectrum as 2.90 MeV and a higher average energy of 3.85 MeV for the isolated PuBe spectrum. The average neutron spectra energies were again used for the final experiment with three sources, which included WGPu, to discriminate a $^9\text{Be}(\alpha,n)$ neutron spectrum from a Watt fission neutron spectrum. The AmBe had an average energy of 3.85 MeV, which was higher than the average energies of the WGPu and ^{252}Cf spectra of 3.23 MeV and 3.20 MeV respectively. Due to the similar shape and average energies of the WGPu and ^{252}Cf neutron spectra, the isolated gamma-ray spectra were needed to discriminate the two sources. The isolated WGPu spectrum showed two prominent peaks at 0.38 MeV and 0.64 MeV which are consistent with decay energies from WGPu. The isolated ^{252}Cf spectrum did not feature these prominent peaks but was consistent with a gamma-ray spectrum from fission.

Because the isolated spectra are subject to resolution effects imparted by detection and the reconstruction process, unfolding methods will be investigated so that the isolated spectra will better match the emitted source spectra.

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