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# Spectra Observed following Cargo Irradiation

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***Abstract*— We present calculations of photon spectra observed following irradiation of bare HEU, HEU embedded in steel and wood cargos, and steel and wood alone. These spectra might be useful starting points for statistical detection efforts aimed at determining whether fissile material is present in a cargo. Detailed comparisons between calculations and experiments are presented and overall quite good (small $\chi^2$) agreement is found. We do not present a complete solution to the problem of determining whether a given spectrum contains contributions from post-fission photons. However, it is shown that a brute-force fitting of observed spectra in terms of a few calculated "basis" spectra gives meaningful predictions about the presence of $^{235}$U in cargo. Though this may not be the most powerful method, it does give well defined confidence limits and seems to have strong predictive power.**

***Index Terms*— Cargo Interrogation, Signal Analysis**

## I. Introduction

The basic goal of cargo interrogation is to estimate the probability that a given cargo contains fissile material. There are two parts to this estimate for interrogation techniques that rely on observing post-fission $\beta$-delayed photons. Very roughly, one of these can be viewed as setting the magnitude of the signature of fissile material. This involves the strength of the interrogating neutron source, the mass of the fissile material, the attenuation of incident neutrons and outgoing photons, and the time-dependent spectral distributions produced in the photon detectors. With the exception of the latter, these aspects have been very well studied in a simulation campaign spearheaded by M.-A. Descalle and J. Hall. The second part involves finding some method for quantifying the contribution of post-fission photons to observed spectra.

So far we have not found a way to take an observed spectrum and specify the presence (or absence) of fissile material with a well defined probability. The initial observation made by Norman and Prussin [1] - that post-fission photons are characterized by a long-lived high-energy component - has been fruitfully used as a starting point. However, fitting the time decay constants characterizing evolution of high energy photons makes use of only a small part of a spectrum. It seems that more powerful approaches are possible. To this end we have undertaken an effort to build machinery capable of predicting observed spectra for different sources and irradiation/counting histories. It is hoped that reliable calculations of observed spectra will enable use of a broad class of tools to search for signatures of fission. To illustrate this we present a quantitative and seemingly useful method for inferring the presence or absence of fissile material from observed spectra.

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## II. Overview of the Method

Here we describe our approach for predicting observed spectra. In practice cargos are irradiated for a finite period of time, perhaps many times in succession, and complicated photon collection histories are acquired. The code we've written and all of the calculations we present correctly incorporates such irradiation histories in a straightforward way. For the purpose of simplifying the description of our method, we consider the case of a $\delta$-function irradiation at time $t = 0$. This irradiation could produce $^{16}$N by an (n,p) reaction, fission fragments, or any number of other interesting unstable nuclides.

The basic quantity we want to calculate is

$$P(E_d, t) \tag{1}$$

the probability that following production of a radionuclide, our detector records an event of energy $E_d$ at time $t$ due to the emission of a photon with known absolute intensity per decay. Several things go into this calculation. First, we must know the photon source function - or probability that the unstable particle emits photons of energy $E$ at time $t$:

$$s_\gamma(E, t). \tag{2}$$

For single nuclides this probability is calculated from the half-lives and decay schemes found in the Table of Isotopes. For fission fragments this probability is calculated as described in [2]. That calculation includes a detailed account of evaluated fragment distributions as well as experimentally-determined decay properties of some 2000 isotopes.

Next we need to account for the influence of scatterings suffered by photons on their way to the detector. Formally this can be written as a function

$$C(E, E') \tag{3}$$

giving the probability that a photon emitted with energy $E$ arrives at the detector with energy $E'$. In general it seems that $C$ could depend in a complicated way on how cargo is loaded, the location and physical surroundings of the detectors, etc. For the present purposes, we have decided to use a simple parametrization to represent all of these. Specifically, we suppose that some fraction $f_C$ of the photons undergo one Compton scattering through an angle less than 90 degrees before arriving at the detector. Though it is shown in the next section that this seems to work well, it isn't yet clear if a more sophisticated account of Compton scattering will be needed. Our simple prescription certainly misses important effects such as the energy dependence of photo-atomic interaction cross sections. One more realistic possibility is to calculate $C$ from

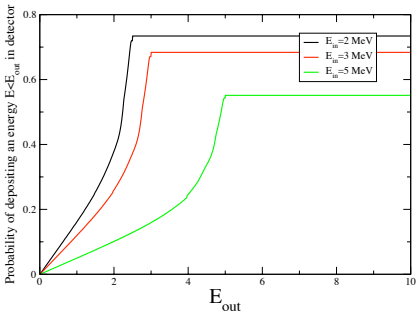


Fig. 1. Cumulative probability distributions characterizing energy deposition in a $2' \times 2' \times 6''$ plastic scintillator. Here $E_{in}$ is the energy of the photon entering the detector and $E_{out}$ is the energy deposited in the detector. The different distributions have been multiplied by a common factor.

simulations of photons transported through different thicknesses of material.

The last ingredient in our mix accounts for the response function of the detector, or probability that a photon entering the detector with energy $E$ is observed as an event of energy $E_d$:

$$D(E, E_d). \tag{4}$$

Because it is hard to calculate $D$ we have broken it into two separate parts:

$$D(E, E_d) = D_{E_d} \times D_S. \tag{5}$$

Here $D_{E_d}$ is the probability that a photon of energy $E$ deposits an energy $E_d$ in the detector and $D_S$ is a smoothing function. The virtue of this approach is that $D_{E_d}$ can be calculated from Monte Carlo simulations while cogent guesses are expected to reproduce the smoothing functions.

We have taken calculations of $D_{E_d}$ from work by Marie-Anne Descalle. Figure 1 shows cumulative probability distributions calculated for a $2' \times 2' \times 6''$ plastic scintillator. It is noted that the probability for full energy deposition is very small, but the probability for a photon to deposit $90\%$ of it's energy is appreciable - around 1/2. The smoothing function $D_S$ has been approximated as a Gaussian with standard deviation $\sigma_E = 0.4\text{MeV}\sqrt{E/6\text{MeV}}$.

With the above ingredients we can write

$$P(E_d, t) = s_\gamma(E, t) C(E, E') D(E', E_d). \tag{6}$$

Spectra are calculated through Monte Carlo sampling of eq. 6. Spectra involving complicated irradiation/counting histories were calculated by making use of appropriate translations in time.

## III. Comparisons between calculated and observed spectra

In this section we compare calculated and observed spectra. Some important features of different spectra are also pointed out. No attempt has been made to include fine details of the experimental setup in the calculations. Our hope is that a simple prescription will suffice for accurate predictions of spectra observed following interrogation of many different cargos.

Experimental spectra were collected following irradiation of bare HEU and HEU embedded in wood and steel assemblies. As well, spectra were observed following irradiation of wood and steel in the absence of HEU. To get a quantitative estimate of how well our calculations reproduce experiments, we fit each observed spectrum $S(E, t)$ with a calculated spectrum

$$C(E, t) = \sum_i c_i f_i(E, t). \tag{7}$$

Here the $f_i$ are the different "basis" spectra corresponding to our calculations for: (i) the spectrum $F$ of post-fission $\beta$-delayed photons, (ii) the spectrum $N$ of photons emitted following $\beta$ decay of $^{16}$N, (iii) the spectrum $M$ of photons emitted following $\beta$-decay of $^{56}$Mn and (iv) a background spectrum $B$. By definition the background spectrum is independent of time. We estimated this from the spectrum observed at late times: $B(E) \equiv S(E, t = 90\text{sec})$. Other basis spectra were calculated as described in the previous section.

The coefficients $c_i$ in eq. 7 are independent of time and energy and are found by minimizing

$$\chi^2 = \sum_{E_j} \sum_{t_i} \left( \frac{C(E_i, t_j) - S(E_i, t_j)}{\sigma_{ij}} \right)^2. \tag{8}$$

Careful attention should be paid to choosing the range in energy and time over which $\chi^2$ is calculated. In the next section we discuss how this choice influences predictions about the presence of fissile material. For the purposes of this section we wanted to present a more-or-less global account of how well calculations reproduce experiments. For this reason we chose to first fit the data over a large range in energy and time: $E_i = 1.5, 1.52, 1.54, 1.56, ....5.96, 5.98, 6.$ MeV and $t_i = 3, 4, 5, ...88, 89, 90$ sec. The uncertainty appearing in eq. 8 is estimated as

$$\sigma_{ij}^2 = S(E, t) + \left( \frac{dS}{dE} \sigma_E \right)^2 + 1. \tag{9}$$

The first term on the right hand side of this equation just represents Poisson counting statistics. The second term is an attempt to incorporate our uncertainty in the detector gain and to minimize the influence of miscalculations involving very rapid changes in the energy dependence of the spectrum. We took $\sigma_E = 60$ keV, which probably underestimates uncertainties in the energy dependence of the detector response.

Figures 2, 4 and 6 show calculated and observed spectra corresponding to irradiation of steel, wood and HEU. In each figure results for an early time and late time are shown. It should be emphasized that the fits were done using eq. 8 and not for each time separately. Figures 3, 5 and 7 show the relative contributions of different sources to the total calculated spectra. Note that because $^{56}$Mn is so long-lived ($\tau \approx 2.6$ hours) decay of this nuclide is largely accounted for by the background we have defined. For this reason one can't get an idea about how much steel is present in a cargo with our method.

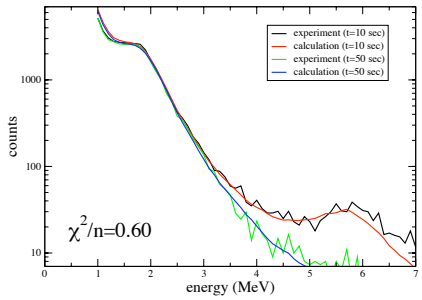


Fig. 2. Comparison between observed and calculated spectra for irradiation of steel. Results for an early time (t=10 sec) and late time (t=50 sec) are shown. The ratio of $\chi^2$ to the number of data points used in the fit ($n \approx 20,000$) is 0.60.

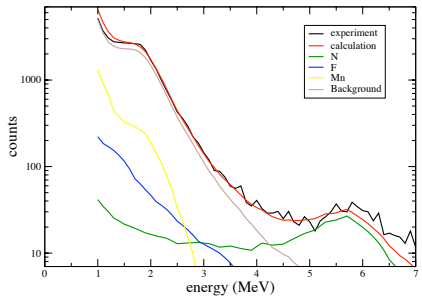


Fig. 3. Experimental and calculated spectra for irradiation of a steel cargo and counts collected 10 sec following the end of irradiation. The total calculated and experimental counts shown are the same as those shown in the previous figure. In addition, contributions of different calculated spectra corresponding to (N) nitrogen decay, (F) post-fission photons, (M) manganese decay and a time-independent background (B) are shown.

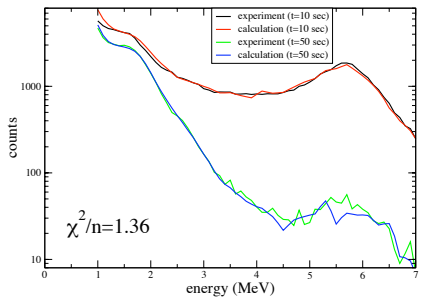


Fig. 4. Comparison between observed and calculated spectra for irradiation of wood. Results for an early time (t=10 sec) and late time (t=50 sec) are shown. The ratio of $\chi^2$ to the number of data points used in the fit is 1.36.

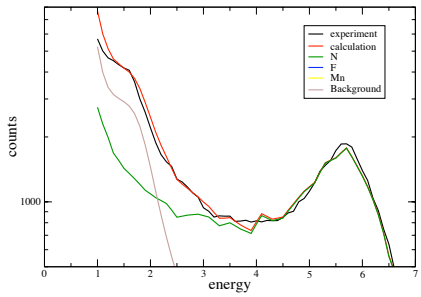


Fig. 5. Experimental and calculated spectra for irradiation of a wood cargo and counts collected 10 sec following the end of irradiation. The total calculated and experimental counts shown are the same as those shown in the previous figure. In addition, contributions of different calculated spectra corresponding to (N) nitrogen decay, (F) post-fission photons, (M) manganese decay and a time-independent background (B) are shown.

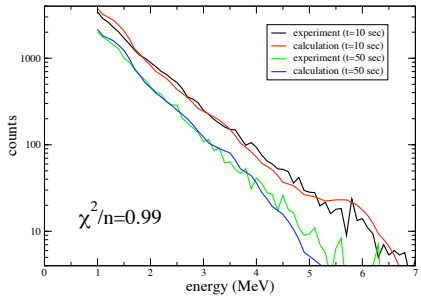


Fig. 6. Comparison between observed and calculated spectra for irradiation of HEU. Results for an early time (t=10 sec) and late time (t=50 sec) are shown. The ratio of $\chi^2$ to the number of data points used in the fit is 0.99.

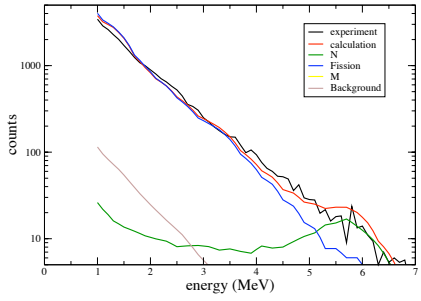


Fig. 7. Experimental and calculated spectra for irradiation of an HEU cargo and counts collected 10 sec following the end of irradiation. The total calculated and experimental counts shown are the same as those shown in the previous figure. In addition, contributions of different calculated spectra corresponding to (N) nitrogen decay, (F) post-fission photons, (M) manganese decay and a time-independent background (B) are shown.

The ratio of $\chi^2$ to the number of data points used in the fits - $n \approx 20,000$ in the present fits - is shown for each case. All of the $\chi^2/n$ are reasonable, with the largest being 1.4 for the wood irradiation. Overall these fits - with just four free parameters - do a pretty good job of reproducing observed spectra over a large time and energy range.

## IV. A BRUTE-FORCE SOLUTION TO THE DETECTION PROBLEM?

In the previous section we found that functions of the form

$$c_F F + c_N N + c_M M + c_B B \tag{10}$$

can do an excellent job of reproducing observed spectra. To remind the reader we note that $F$ corresponds to the spectrum calculated for post-fission photons, $N$ and $M$ correspond to the spectra calculated for decay of $^{16}$N and $^{56}$Mn and $B$ is the calculated time-independent background. Taking these fits seriously leads us to a straightforward method for determining whether a given observed spectrum shows evidence of fission. Namely, we just fit the observed spectrum and use the inferred value of $c_F$ - along with information about the quality of the fit - to make statements about the presence of fissile material. One nice feature of this approach is that minimizing $\chi^2$ gives formal estimates of the uncertainties in derived parameters. Whether or not the fitted coefficients and their uncertainties have any physical meaning is addressed next.

Table I shows calculated values for $c_F$ and its uncertainty for fits to spectra observed following irradiations of different materials. Results in the column labeled "method 1" were derived using exactly the same fitting approach used in the previous section. This approach involves fitting over a quite broad time and energy range. Of course, we are free to choose the time/energy range over which we sample points used in the evaluation of $\chi^2$. This freedom might be exploited to increase the sensitivity of our fits to the presence of post-fission photons. For example, it may be thought that the photons most characteristic of fission are those emitted with energies around 3 MeV at times less than 10 sec. This suggests we sample points with a bias. Results in Table I under the column labeled "method 2" were calculated with such a bias. For definiteness we chose to sample observed spectra at energy points in the range $1.5 < E < 6$ MeV and with a distribution given by $E_i = E_{i-1} + \delta(E_i)$. Here $\delta(E) = 0.012 + 0.01 \cdot \exp[(E-2.5)^2]$, with $E$ here in units of MeV. Observed spectra were sampled at times between 3 and 60 sec with a bias towards earlier times: $t_i = 1.3 \times t_{i-1}$. Figure 8 shows the distribution of points sampled. The coefficient $c_{\rm M}$ was chosen to be zero for these fits because Mn decay is not expected to contribute appreciably to spectra at energies greater than 2.5 MeV.

As seen from Table I, the method employing a global fit to observed spectra seems to give reasonably good predictions about the presence of HEU. In each case where HEU was embedded in cargo the estimated value for $c_F$ is larger than zero by several standard deviations. The ratios of the $c_F$'s are in quite reasonable agreement with those found from the crude fitting procedure using the rough 6-group model for time dependence of the decay of high-energy fission-product gamma rays. They are also about what one expects from the actual irradiation conditions, although a quantitative analysis is still in progress. Also - the calculated fission coefficient for the wood only cargo is inconsistent with a positive value. However - this method predicts the presence of HEU in the steel only cargo. Though the predicted amount is small, this is still unsatisfactory.

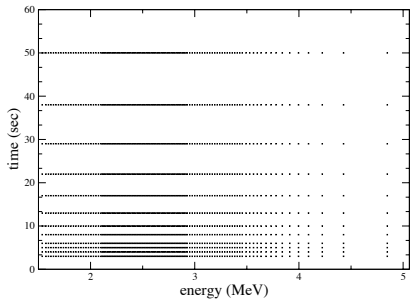


Fig. 8. Time-energy pairs at which observed spectra are sampled in method 2. This choice of points comes from a rough guess about the time/energy most likely to contain interesting information about post-fission photons.

TABLE I

COEFFICIENTS $c_F$ OF THE FISSION CONTRIBUTION TO OBSERVED SPECTRA COLLECTED FOLLOWING IRRADIATION OF DIFFERENT MATERIALS. THE VALUE OF $\chi^2$ CHARACTERIZING EACH EACH FIT IS ALSO SHOWN IN PARENTHESIS. METHOD 1 CORRESPONDS TO A FIT THAT GIVES ROUGHLY EQUAL CONSIDERATION TO ALL TIMES BETWEEN 4 AND 90 SEC AND ALL ENERGIES BETWEEN 1.5 AND 6 MEV. METHOD 2 CORRESPONDS TO A FIT THAT SAMPLES POINTS PREFERENTIALLY IN AN INTERESTING TIME/ENERGY REGION. SEE TEXT FOR MORE DETAILS.

| cargo | method 1 | method 2 |
|---|---|---|
| wood | $-0.0587 \pm 0.0025$ (1.3585) | $-0.1003 \pm 0.0040$ (1.7931) |
| steel | $0.0368 \pm 0.0012$ (0.6026) | $0.0029 \pm 0.0047$ (0.3998) |
| HEU | $0.6694 \pm 0.0017$ (0.9936) | $0.6544 \pm 0.0068$ (1.0413) |
| wood+HEU | $0.2525 \pm 0.0035$ (1.2659) | $0.1190 \pm 0.0183$ (1.0616) |
| steel+HEU | $0.0497 \pm 0.0012$ (0.5955) | $0.0152 \pm 0.0051$ (0.4047) |

Does method 2 - which preferentially samples observed spectra at interesting times and energies - do any better? It seems to for this small sample of irradiated cargos. In each case where HEU is present this method predicts a statistically significant contribution of post-fission photons to the spectra. And in each case where HEU was not embedded in cargo this method predicts a $c_F$ consistent with zero or a negative value. When more experiments are completed it may be found that a simple method like this is broadly useful for predicting the presence or absence of HEU.

Some of the challenges associated with extracting signatures of fission can be seen through consideration of figures 9 and 10. These show contributions of different photon sources to spectra calculated for the wood+heu and steel+heu irradiations. It is seen that at 30 sec following the end of irradiation the

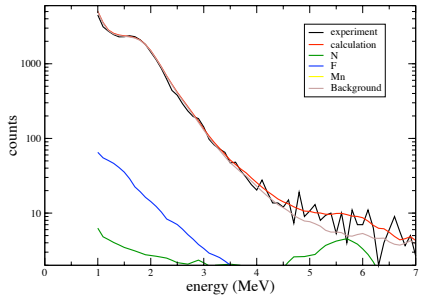


Fig. 9. Experimental and calculated spectra for irradiation of a steel+HEU cargo and photons collected 30 sec after the end of irradiation. The calculational method here is that described as "method 2" in the text. Contributions of different calculated spectra corresponding to (N) nitrogen decay, (F) post-fission photons, (M) manganese decay (not used here but included for consistency with previous plots) and a time-independent background (B) are shown.

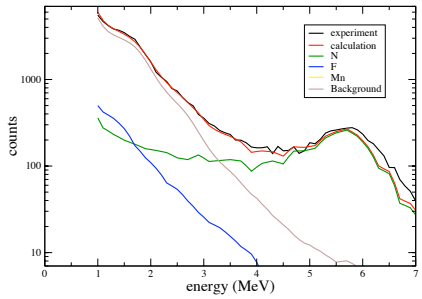


Fig. 10. Same as previous figure except for irradiation of wood+HEU cargo.

predicted contribution of post-fission photons is quite small. In both the wood+heu and steel+heu cases only about 1% of the high energy photons are expected to come from decay of fission fragments.

## V. Summary

We've presented calculations for spectra observed following irradiation of different cargos. These calculations themselves are probably not so useful, but they demonstrate that the machinery needed for accurate predictions of spectra is in a decent state. Comparisons against experiment show statistically meaningful agreement. A simple approach to determining whether or not observed spectra indicate the presence of fissile material was also presented and found to be promising.

## References

[1] Norman, E. B. et al. 2004, Nucl. Instrum. Methods Phys. Res. A 521, 608
[2] Pruet, J., Hall, J., Descalle, M.-A. & Prussin, S.G. 2004, Nucl. Instrum. Methods Phys. Res B, 222, 403