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Validation and uncertainty quantification of detector response functions for a 1″×2″ NaI collimated detector intended for inverse radioisotope source mapping applications

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ABSTRACT

Detector response functions (DRFs) are often used for inverse analysis. We compute the DRF of a sodium iodide (NaI) nuclear material holdup field detector using the code named g03 developed by the Center for Engineering Applications of Radioisotopes (CEAR) at NC State University. Three measurement campaigns were performed in order to validate the DRF's constructed by g03: on-axis detection of calibration sources, off-axis measurements of a highly enriched uranium (HEU) disk, and on-axis measurements of the HEU disk with steel plates inserted between the source and the detector to provide attenuation. Furthermore, this work quantifies the uncertainty of the Monte Carlo simulations used in and with g03, as well as the uncertainties associated with each semi-empirical model employed in the full DRF representation. Overall, for the calibration source measurements, the response computed by the DRF for the prediction of the full-energy peak region of responses was good, i.e. within two standard deviations of the experimental response. In contrast, the DRF tended to overestimate the Compton continuum by about 45–65% due to inadequate tuning of the electron range multiplier fit variable that empirically represents physics associated with electron transport that is not modeled explicitly in g03. For the HEU disk measurements, computed DRF responses tended to significantly underestimate (more than 20%) the secondary full-energy peaks (any peak of lower energy than the highest-energy peak computed) due to scattering in the detector collimator and aluminum can, which is not included in the g03 model. We ran a sufficiently large number of histories to ensure for all of the Monte Carlo simulations that the statistical uncertainties were lower than their experimental counterpart's Poisson uncertainties. The uncertainties associated with least-squares fits to the experimental data tended to have parameter relative standard deviations lower than the peak channel relative standard deviation in most cases and good reduced chi-square values. The highest sources of uncertainty were identified as the energy calibration polynomial factor (due to limited source availability and NaI resolution) and the Ba-133 peak fit (only a very weak source was available), which were 20% and 10%, respectively.

1. Introduction

A detector response function (DRF) is a function that converts the energy-dependent flux of incoming source particles incident on a detector into a detector response (pulse height) spectrum corresponding to that observed in experimental detector measurements. The DRF is used to characterize an unknown source distribution as in the problem of nuclear material controls and accountancy in quantifying Material Unaccounted For in the form of holdup [1], the target application of this work. DRFs have been investigated and developed for several research and industrial detection applications. These applications include neutron depth profiling in substrate manufacturing [2], cosmic radiation detection and atmospheric monitoring [3], and positron emission tomography [4]. DRFs have been proposed for nuclear safeguards and security applications as well, such as border monitoring for illegal transport of radioactive materials, cargo and package monitoring, and unknown source identification at source recovery sites.

At present, neither a rigorous mathematical formulation nor a complete physical model exists to describe DRFs. Instead, several stochastic (Monte Carlo) and empirical models are available and

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reported in the literature. Gardner developed a DRF model through empirical curve fitting and Monte Carlo analysis [5]. He validated his DRF against high-fidelity experimental measurements reported by Heath [6] for 3″×3″ and 6″×6″ bare NaI detectors using Cs-137 sources centered on the detector's front axis at a distance of 10 cm. There was agreement between Gardner's DRF-computed responses and Heath's benchmark detector measurements of the same detector sizes within two standard deviations of the measured Poisson uncertainty. Gardner's model was also found to be more efficient (required far fewer particle histories for comparatively accurate calculation) and was shown to match better with the Heath experiments than MCNP's F8 pulse-height tally.

In this work the NaI DRF model developed by Gardner is used to characterize a NaI 1″×2″ detector for on-axis, off-axis, and attenuated configurations and to validate it against experimental measurements using Cs and HEU sources. Also, uncertainty in the model is calculated by both Frequentist and Bayesian methods and compared to measurement and Monte Carlo transport uncertainties.

1.1. Detector response functions

Mathematically, the DRF, denoted $R(H, E)$, is defined as the probability that a photon incident on the detector with energy E yields a pulse with height H [7]. We employ Gardner's DRF model in our work due to its efficiency and also because MCNP simulates responses according to direct energy deposition in the detector crystal without generating a DRF. A DRF comprises a matrix whose rows represent the energy of an incident photon and columns correspond to detector channels. Elements of the matrix indicate the probability of producing a pulse in a channel due to an incident photon with the given energy. DRFs have been employed in various applications and more recently have gained interest for use in inverse transport problems. An accurate DRF matrix equipped with uncertainty estimates is essential for the success of these applications. The reason for this is the fact that inverse problems seeking the determination of a radiation source distribution from a set of measured detector responses typically require repeated evaluation of modeled detector responses in the process of searching the state space for an optimal inverse solution. For computationally intensive models like radiation transport, repeating such forward computations for each evaluated state would be prohibitively expensive. Instead, computing the adjoint flux (particle importance) for the same configuration using the column sum of the detector's response function matrix as the adjoint source vector provides an inexpensive means to evaluate the response as the inner product of the resulting adjoint flux with the source distribution characterizing the tested state. Consequently, more states can be tested in search for the distribution that best fits the measured responses, thus improving the quality of the solution to the inverse problem.

Gardner's model generates a DRF for a desired detector size, source distance and source energy (single peak), and it accounts for the nonlinear dependence of NaI scintillation efficiency $\left(\frac{scintillation\ light\ yield}{energy\ deposited}\right)$ [%] on the energy deposited in the detector by the incident photon through the following steps [5]. First, a Monte Carlo calculation is conducted with the DRFNCS code [8] to simulate several hundred detector response spectra where photons interactions are only allowed to occur within the detector cell (forced collisions), but leakage of secondary particles is allowed, producing the continuum segment of the spectra. Only about 100,000 particle histories are necessary to produce results with uncertainty under 1%, whereas MCNP F8 Gaussian energy-broadened (GEB) spectra require at least 100 times as many histories to post-process the Gaussian spectral peaks.

Next, the peaks are stripped from the response spectra so that each continuum can be processed separately. Principal component analysis (PCA) is then performed on the correlated response variables and the covariance matrix to produce a small set of uncorrelated variables (principal components). The principal components and the mean vector are stored as data that reproduces an accurate continuum when multiplied with the desired number of channels. Essentially, the continuum can be constructed efficiently without the need for repeated Monte Carlo simulations for each DRF generated.

So, when a new DRF is to be generated, the algorithm need only to generate the full-energy peak of interest by Monte Carlo transport simulation and adds this contribution to the archived continuum to produce the desired DRF [8]. The modified version of DRFNCS (adjusted by the nonlinear scintillation efficiency) is implemented in the computer code g03.

Finally, the Monte Carlo simulation in g03 is modified by several empirical equations to correct pieces of the spectra that are not simulated fully by the Monte Carlo calculation. The g03 DRF peak section is spread according to the power law based on Gaussian peak fitting of measured detector response spectra

$$\sigma_T(E_I) = aE_I^b, \tag{1}$$

where a and b are empirical fit parameters, and E_I is the energy of the incident photon. $\sigma_T(E_I)$ is the Gaussian uncertainty of the measured full-energy peak. Equation (1) is a semi-empirical model representing the Gaussian peak response spread, whose parameters are found by the least-squares fitting of standard deviations of experimentally measured full-energy peak responses produced by the detector of interest [5].

The flat Compton continuum of the DRF is computed by various empirical fits of the entire experimental responses (not only the peaks). This is necessary because there is as of yet some unmodeled phenomena causing a higher magnitude of the simulated continuum than predicted by the included physics models and observed in the measured data. Simple Compton scattering and partial energy deposition physics due to electron or photon leakage through the detector walls can predict the general flat shape of the Compton continuum but underestimate its magnitude. A normalization factor was developed to account for this effect called the electron range multiplier, since the effect causing the underestimation of the continuum was believed to be connected to the electron range in crystalline materials [9]. The empirical relation is given by

$$R_e = 1 + A_1 e^{(-A_2 E_I)} + A_3 e^{(-A_4 E_I)} \tag{2}$$

$$A_1 = 39.662, \quad A_2 = 3.4052, \quad A_3 = 1.5434, \quad A_4 = 0.1576,$$

where E_I is again the energy of the incident photon, and A_1 through A_4 are fit parameters determined from experimental responses. This unitless factor (R_e) is a pseudo-electron range factor designed to correct the magnitude of the synthetic Compton continuum produced by Gardner's DRF. It was originally fit through trial and error for uncollimated 3″×3″ NaI detectors [9] and therefore may be a source of additional error for the 1″×2″ NaI detector of interest in this work.

1.2. Uncertainty quantification

In the process of comparing measured to computational model results there is measurement uncertainty, model uncertainty, and numerical (simulation) uncertainty. Radiation counting (measurement) and Monte Carlo sampling uncertainty approximately follow a Poisson distribution [10,11].

Determination of the model parameter uncertainty is a more difficult task. Since the core of Frequentist Theory requires a large