Nuclear Engineering and Technology xxx (2017) 1–4

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Original Article

Neutron and gamma-ray energy reconstruction for characterization of special nuclear material

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ARTICLE INFO

Article history:
Received 25 May 2017
Accepted 18 June 2017
Available online xxx

Keywords:
Neutron spectroscopy
Spectrum unfolding
Neutron imaging

ABSTRACT

Characterization of special nuclear material may be performed using energy spectroscopy of either the neutron or gamma-ray emissions from the sample. Gamma-ray spectroscopy can be performed relatively easily using high-resolution semiconductors such as high-purity germanium. Neutron spectroscopy, by contrast, is a complex inverse problem. Here, results are presented for ^{252}Cf and PuBe energy spectra unfolded using a single EJ309 organic scintillator; excellent agreement is observed with the reference spectra. Neutron energy spectroscopy is also possible using a two-plane detector array, whereby time-of-flight kinematics can be used. With this system, energy spectra can also be obtained as a function of position. Spatial-dependent energy spectra are presented for neutron and gamma-ray sources that are in excellent agreement with expectations.

1. Introduction

Energy spectroscopy is useful for characterizing special nuclear material (SNM). Isotope-specific gamma rays are emitted at specific energies, which allows for very accurate determination of the SNM composition. Semiconductors and inorganic scintillators are the most commonly used detectors for these applications.

Neutron spectroscopy, by contrast, is a much more difficult task. The response of organic scintillators is related to the incident neutron energy; however, neutrons interact in organic scintillators through elastic scattering, which creates a weak coupling between the measured detector response and the energy spectrum of the incident neutrons.

If there are multiple sources in field-of-view of the detector, imaging techniques can be used to isolate the location of the individual sources. Once the location of the sources is determined, the energy spectrum of the incident particles may be extracted for any region in the field-of-view using the coincident scatter data from the imager.

Here, we present neutron energy spectrum unfolding results from a single EJ-309 liquid scintillator. In addition, we present spatial-dependent energy spectra using our dual-particle imaging array of EJ-309 and NaI(Tl) scintillators.

2. Neutron energy spectrum unfolding

Neutron energy spectra can be derived by unfolding the detector response from the detected pulse height spectrum. In general, the unfolding problem can be defined as follows:

$$z_{0i} + e_i = \sum_{j=1}^{M} R_{ij}\Phi_j \tag{1}$$

where z_{0i} is the light output in the ith bin; e_i is the error associated with the ith light output bin; M is the number of detection channels; $R_{ij}(E)$ is the detector response element for the ith light output bin and the jth neutron energy bin; $\Phi_j(E)$ is the Neutron spectrum flux cm^{-2}, in the jth energy bin.

2.1. Solution methodology

One approach to solving Eq. (1) is direct inversion of the response matrix. The matrix itself can be obtained by simulating monoenergetic neutrons in the energy range of interest, using a Monte Carlo code such as MCNPX-PoliMi and MPPost [1–3]. Because of the ill-conditioning of the response matrix, the direct inversion approach typically yields an oscillatory solution for the incident energy spectrum; this solution is not stable with respect to the uncertainty of the measured data. The statistical fluctuations and the amplitude of the oscillations can be as large as the estimated values themselves.

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http://dx.doi.org/10.1016/j.net.2017.06.005

To overcome this issue, regularization is often applied. One type of regularization function is based on spectral entropy. It can be shown that the entropy is a measurement of the smoothness of a histogram [4]. Applying a regularization function consists in imposing a smoothing condition to the estimate of the flux histogram. A regularization function, as in Eq. (2), is obtained using a Bayesian approach, which incorporates the prior information.

$$H(y) = \sum_{i=1}^{M} p_i \log p_i \tag{2}$$

$H(\boldsymbol{y})$ is the entropy of a probability distribution $\boldsymbol{p} = (p_1, \ldots, p_n)$; the values to be estimated, $\boldsymbol{y}$, are random variables. According to the Bayes theorem, the joint probability density function, $f(\boldsymbol{y}|\boldsymbol{z})$, depends on $L(\boldsymbol{z}|\boldsymbol{y})$, as in Eq. (3). $L(\boldsymbol{z}|\boldsymbol{y})$ is the likelihood function, i.e., the conditional probability for the data $\boldsymbol{z}$, given $\boldsymbol{y}$ and the *a priori* information $ph(\boldsymbol{y})$.

$$f(y|z) \propto L(z|y)ph(y) \tag{3}$$

One wants to find $\hat{y}$ that maximizes $f(\boldsymbol{y}|\boldsymbol{z})$, or its logarithm. In the absence of any other *a priori* information, the probability density function $f(\boldsymbol{y}|\boldsymbol{z})$ of a certain histogram is proportional to the number of possible combinations of the data in the bins, i.e., the entropy of the data.

2.2. Results

We evaluated ^{252}Cf and PuBe neutron spectra obtained using the MAXED code, a Bayesian algorithm based on entropy maximization [5]. This method translates prior knowledge available about the problem (such as spectra positivity and energy boundaries) into a prior model, which is coupled with the observation model (or likelihood) to derive the posterior distribution of the unknown neutron spectra.

Experimental data were collected for comparison. One 7.62 cm (diameter) × 7.62 cm (length) EJ-309 detector (Eljen Technology, Sweetwater, TX, USA) was irradiated with two sources—^{252}Cf (~25 μCi) and PuBe (1 Ci)—to evaluate its performance as neutron spectrometer. Source-to-detector distance was 1.5 m for ^{252}Cf and 1 m for PuBe. Prior to neutron irradiation, the detector was calibrated using a ^{137}Cs source, to match 80% of the Compton edge peak (478 keVee) to a pulse amplitude of 0.3 V. The neutron spectrum was separated from the photon spectrum using a pulse shape discrimination algorithm, based on charge integration [6].

Figs. 1 and 2 show the unfolded spectra from ^{252}Cf and PuBe sources. In Fig. 1, the peak in the unfolded spectrum appears at higher energies than the reference because of the detection threshold. Given that, we consider the unfolded spectra in satisfactory agreement with the reference spectra, i.e., the analytic model by Mannhart [7] for spontaneous fission ^{252}Cf source and the measured one for (alpha, n) PuBe source [8]. A low energy limit of about 400 keV and 1 MeV was achieved for PuBe and ^{252}Cf, respectively. Although the simulated response matrix extends below 100 keV, the practical unfolding limit at low energies is a few hundred keV because of uncertainties in the light output function and fidelity of the pulse shape discrimination. The ^{252}Cf spectrum was unfolded using a flat guess spectrum; conversely, the reference spectrum was incorporated as prior information, to unfold the PuBe data. Ill-conditioning of the EJ-309 response matrix and the smoothing entropy maximization did not allow to resolve the peaks in the spectrum, owing to the excited states of ^{12}C [^{9}Be(alpha, n)^{12}C*], when a flat guess spectrum was used.

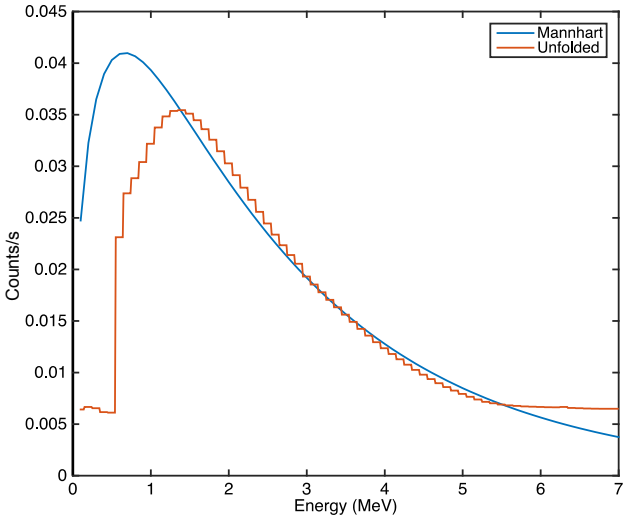

Fig. 1. Unfolded neutron energy spectrum from ^{252}Cf detected with an EJ-309 liquid scintillator; the Watt spectrum from Mannhart [7] is shown for reference.

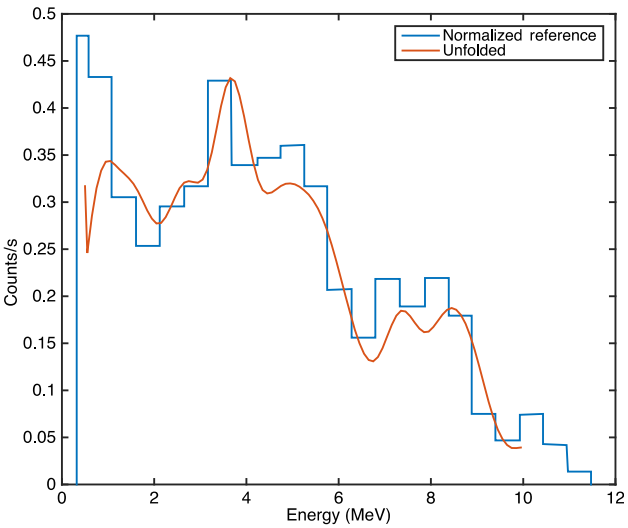

Fig. 2. Unfolded neutron energy spectrum from PuBe detected with an EJ-309 liquid scintillator; a normalized PuBe spectrum from Knoll [8] is shown for reference.

3. Spatial-dependent energy spectra

The dual-particle imager (DPI) is a combined Compton and neutron scatter camera that is sensitive to both neutrons and gamma rays [9, 10]. The system works as an imager and spectrometer for both particle types. The system is configured in two planes and uses liquid organic and NaI(Tl) scintillators. The pulse shape discrimination capabilities and fast response of the liquid scintillators make them well suited for this system.

Image reconstruction was performed on the data using the stochastic origin ensemble method. This method uses Markov chain Monte Carlo to iteratively construct an image with improved signal/noise ratio and resolution compared to backprojection imaging [11]. The incident energy is obtained for both neutrons by using time-of-flight, and gamma-rays by summing both energy depositions. Spectra are isolated by using the stochastic origin ensemble algorithm to create separate images binned by energy. The intensity of a particular pixel can then be translated from each energy binned image to an energy spectrum. To maintain adequate

Please cite this article in press as: S.D. Clarke, et al., Neutron and gamma-ray energy reconstruction for characterization of special nuclear material, Nuclear Engineering and Technology (2017), http://dx.doi.org/10.1016/j.net.2017.06.005