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Fast neutron spectrum unfolding for nuclear nonproliferation and safeguards applications

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Summary. — We present new neutron spectrum unfolding results obtained from measurements with Cf-252 and plutonium-oxide sources. The precise knowledge of the neutron energy spectrum provides information about the presence or absence of fissile material and about the characteristics of the material. We used a neutron spectrum unfolding technique based on a modification of the least-squares method. The main innovation is the use of a Krylov subspace iteration which performs better on ill-conditioned systems of linear equations than standard direct-solution methods. The proposed technique performed well in the unfolding of measured neutron pulse-height distributions from a Cf-252 neutron source and from plutonium-oxide samples and could be easily implemented in a portable neutron spectroscopy system for nuclear nonproliferation and safeguard applications.

PACS 29.30.Hs – Neutron spectroscopy.
PACS 29.40.Mc – Scintillation detectors.
PACS 25.85.Ca – Spontaneous fission.

1. – Introduction

Neutron measurements based on the unfolding of the incident neutron energy spectrum have been performed often in the past decades. Early attempts used Bonner spheres, which provide an estimate of the neutron energy spectrum by performing measurements with spheres of various diameters. More sophisticated measurements unfold the incident neutron spectrum by analyzing the pulse height distributions measured using liquid-scintillation detectors [1-4]. The evaluation of the uncertainties associated with various unfolding methods has also been the subject of recent investigations [5].

More recently, applications in nuclear nonproliferation and safeguards have led to a renewal of interest in these unfolding procedures and in their application to radiological

sources and bulk nuclear materials. In a previous paper [6], we describe a new method to perform neutron energy spectrum unfolding based on a modification of the least-squares approach called sequential least-squares (SLS) method. The SLS technique makes use of Krylov subspaces for the solution of the minimization of the unfolding problem. This method was shown to be more robust to noise when compared to a simple least-squares approach [6].

In this paper, we present new results from the application of the SLS method to experimental data recently obtained at the JRC-Ispra, Italy on plutonium oxide samples and from a Cf-252 source. We show that the unfolded spectra agree reasonably well with published data in the literature. Moreover, we show that the plutonium oxide data can be distinguished from the Cf-252 data, a capability that is of particular interest in nonproliferation applications.

2. – Formulation of the unfolding problem

In general, unfolding procedures with liquid scintillators can be seen as a mapping from the n-dimensional space of the detector response to the m-dimensional space of the neutron energy flux [4]. The detector response, the detector count rates, and the incident neutron spectrum are related through the Fredholm integral equation of the first kind,

$$N(L) = \int R(E, L)\Phi(E)\mathrm{d}E, \tag{1}$$

where E is the neutron energy, L is the measured light output (or pulse height), $N(L)$ is the count rate density corresponding to L, and $R(E, L)$ is the detector response matrix. Obtaining the energy distribution of the incident neutrons, $\Phi(E)$, from the measured detector pulse height distribution amounts to solving an ill-posed inverse problem, for which: i) the solution is not unique and ii) the solution(s) do(es) not depend continuously on the data.

Upon discretization, eq. (1) reads

$$N_i = \sum_{j=1}^{m} R_{ij}x_j, \quad \text{for } i = 1, \ldots, n, \tag{2}$$

where N_i is the binned count rate corresponding to a certain interval of the measured light output (pulse height) in the i-th channel, x_j is the incident neutron fluence in the j-th energy group, and R_{ij} is the corresponding element of the response matrix. In the response matrix, each row corresponds to a given neutron energy and each column corresponds to a given pulse height. The following matrix and vectors notation will be used hereafter:

$$N = \begin{bmatrix} N_1 \\ \vdots \\ N_n \end{bmatrix}, \qquad x = \begin{bmatrix} x_1 \\ \vdots \\ x_m \end{bmatrix}, \qquad R = \begin{bmatrix} R_{1,1} & \cdots & R_{1,m} \\ \vdots & \ddots & \vdots \\ R_{n,1} & \cdots & R_{n,m} \end{bmatrix}.$$

3. – Detector response matrix

Monte Carlo simulations were used to generate the response matrix R_{ij} for the liquid-scintillation detectors used in this work. Previous work has shown that the MCNP-PoliMi

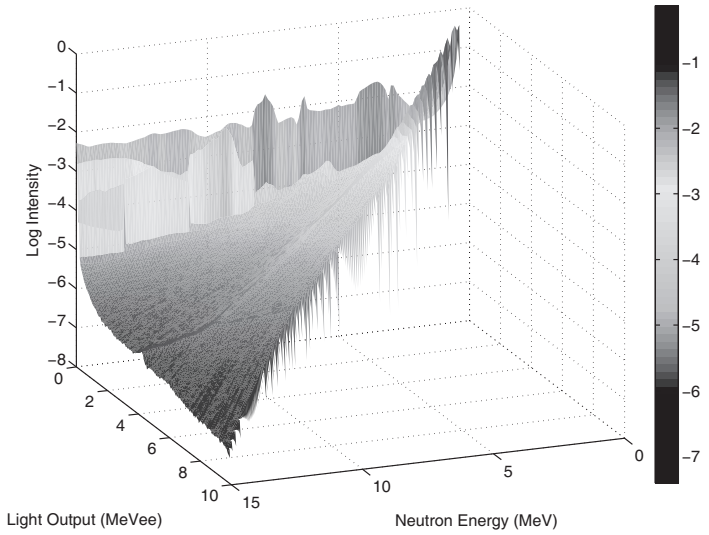

Fig. 1. – Response matrix from MCNP-PoliMi simulation.

code [7] can be used to accurately calculate the detector response matrix. This method is faster and more practical than measuring the detector response to monoenergetic neutrons at various energies.

To simulate a complete detector response matrix in the energy range of interest, we considered monoenergetic neutrons from a surface source emitting perpendicularly to the front face of a cylindrical liquid-scintillator detector, having the same dimensions as the one used in the experiments (see sect. **5**). Subsequently, we used a specialized algorithm for the analysis of the interactions occurring in the scintillator material. This analysis takes into account neutron scattering on hydrogen, neutron scattering on carbon, and secondary photons that may be generated on carbon when the energy of the incident neutron is above 4.4 MeV. The light output from secondary charged particles produced by neutron reactions within the scintillator is computed using experimentally determined parameters that relate the energy deposited to the scintillator light output. The cumulative effect of multiple scatterings on light output is also taken into account. Further details on this simulation methodology can be found in [7, 8].

The response matrix obtained from MCNP-PoliMi simulation is shown in fig. 1. The resolution of pulse-height bins is 0.010 MeVee and neutron energy group width is 0.020 MeV. From this matrix, a second response matrix was obtained by averaging several pulse height bins and neutron energy groups. This second response matrix was shown to give more stable results in the unfolding procedures. It has pulse-height bins of 0.050 MeVee and neutron energy group width of 0.100 MeV. This matrix was used in the unfolding procedures given in sect. **4**.

4. – Unfolding by the sequential least-squares method

For the practical unfolding problem, errors exist both in the response matrix R, and in the detector response N, which may result in significant errors in the solution vector x. Commonly used unfolding procedures, such as the least-squares method or the direct inversion method, may lead to highly unstable solutions, which suffer from non-physical os-

cillations and negative flux values. For these reasons, a constrained optimization method is used to achieve a physical solution to the unfolding problem.

The constrained optimization problem is defined as

$$\begin{aligned} &\min f(x) = \frac{1}{2}\sum_{i=1}^{n} w_i \left(N_i - \sum_{j=1}^{m} R_{ij} x_j \right)^2 , \\ &\text{subject to } x_j \geq 0 \qquad \text{for} \qquad j \in S_B \equiv \{1, 2, \ldots m\}, \end{aligned} \tag{3}$$

where S_B is the set of indices for the lower bounds, which ensures that fluxes remain non-negative.

This type of constrained optimization problem is most often solved with sequential quadratic programming (SQP) using an active set strategy. At each step of the SQP, a QP subproblem is solved:

$$\begin{aligned} &\min f(x) = \frac{1}{2}\sum_{i=1}^{n} w_i \left(N_i - \sum_{j=1}^{m} R_{ij} x_j \right)^2 \\ &\text{subject to } x_j = 0 \qquad \text{for} \qquad j \in S_A \subseteq S_B. \end{aligned} \tag{4}$$

The constraints are said to be active at x' if x' lies on the boundary of the feasible region. This boundary is formed by the constraints whose indices are members of set S_A, which is referred to as the active set. The set containing members of set S_B that are not in active set S_A is referred to as the inactive set $S_B \backslash S_A$. During the search process, an index may move back and forth between an active and an inactive set.

The constrained optimization problem can then be reduced to the following unconstrained optimization problem:

$$\min f(x) = \frac{1}{2}\sum_{i=1}^{n} w_i \left(N_i - \sum_{j \in S_B \backslash S_A} R_{ij} x_j \right)^2 . \tag{5}$$

This special SQP will be called SLS. The algorithm is described in detail in [6].

5. – Unfolding results

The SLS method was applied to measured pulse height distributions obtained in the setup shown in fig. 2. The photograph shows the detectors used in the measurement system developed at the University of Michigan, which consists of six liquid-scintillation detectors (EJ-309), a 250 MHz, 12 bit digitizer for data acquisition, and analysis algorithms for the determination of neutron pulse height distributions [9, 10].

Measurements were performed on a Cf-252 source and several plutonium oxide samples of varying mass. In the measurements from the plutonium oxide samples, a shield of approximately 2.5 cm of lead was used to reduce the gamma-ray background from the samples. Optimized digital pulse shape discrimination techniques were applied to the measured data from the four detectors placed symmetrically in the horizontal plane surrounding the sample. The data from these four detectors added together to obtain the neutron pulse height distribution needed in the unfolding procedure.

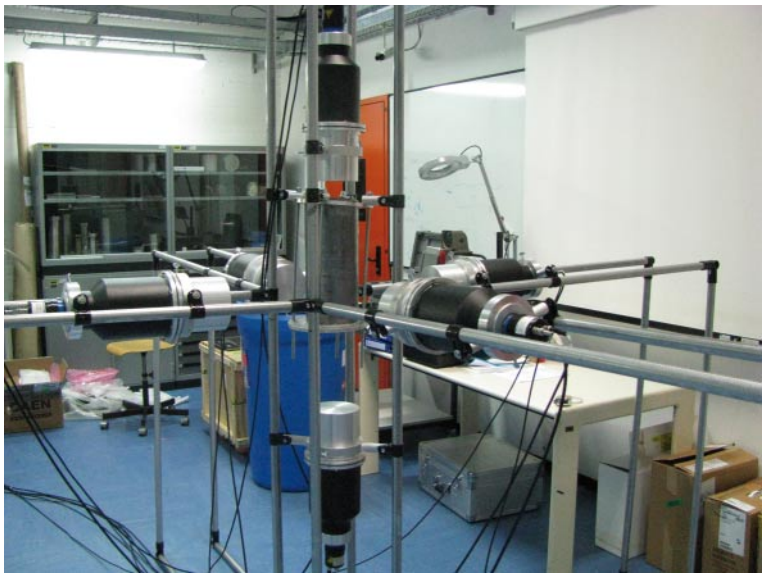

Fig. 2. – Photograph of experimental setup for the acquisition of neutron pulse height distributions (JRC-Ispra, Italy, the August 2008).

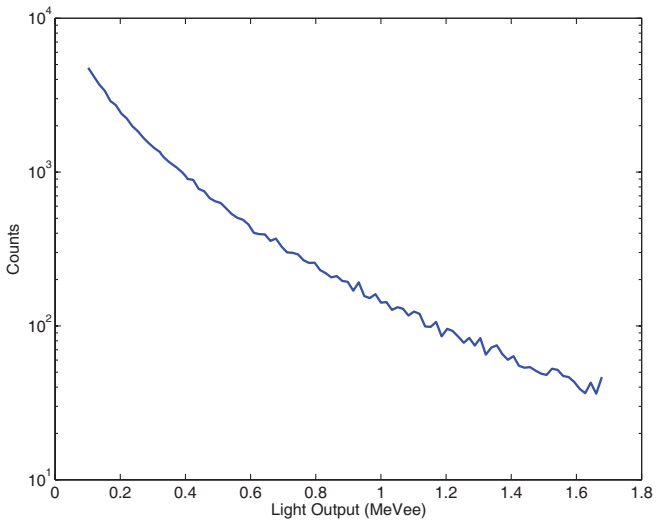

Fig. 3. – Measured neutron pulse height distribution from Cf-252 source.

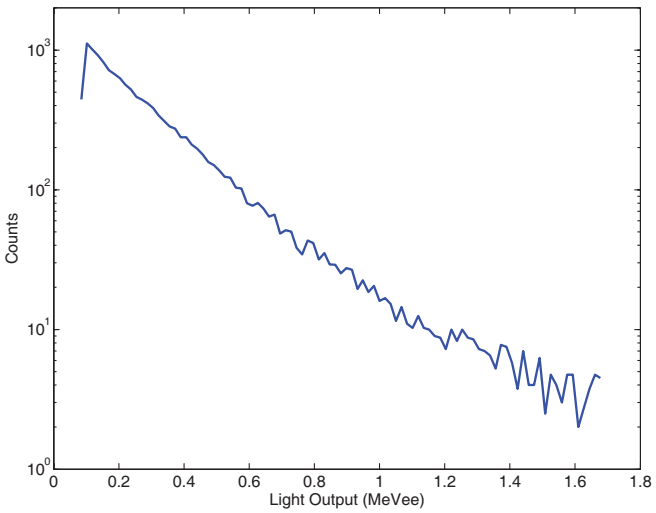

Fig. 4. – Measured neutron pulse height distribution from 100 g plutonium-oxide sample.

Figure 3 shows the neutron pulse-height distributions obtained from the Cf-252 source. Figure 4 shows the neutron pulse-height distributions obtained with a 100 g, low-burnup, plutonium-oxide sample. In the case of the Cf-252 source the neutron production is due solely to the spontaneous fission events, whereas in the plutonium-oxide sample the spontaneous fission of Pu-240, Pu-242, and Pu-238 account for approximately 75% of the neutrons and the remaining 25% originate from (alpha, n) reactions with oxygen. Pu-240, Pu-239, and Pu-238 are the primary contributors of the alpha decays. Consequently, the energy spectrum is harder in the case of the plutonium-oxide sample than in the Cf-252 source in the energy region between 2 and 3.5 MeV [10].

The SLS method was applied to the pulse height distributions shown in figs. 3 and 4.

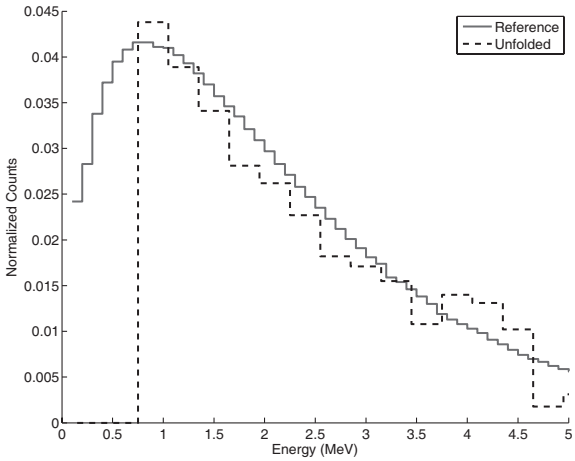

Fig. 5. – Unfolding results for Cf-252 source; the reference spectrum is also shown.

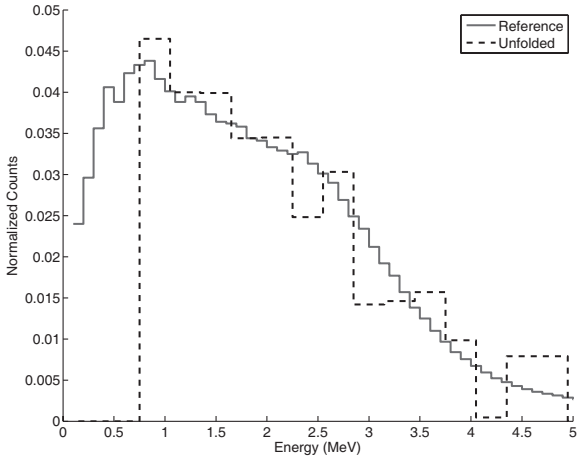

Fig. 6. – Unfolding results for plutonium-oxide source; the reference spectrum is also shown.

The algorithm takes a few seconds to converge (approximately 20 cycles). The resulting neutron energy spectra are shown in figs. 5 and 6.

The neutron spectrum unfolding results for the Cf-252 and plutonium-oxide sample measured data shows good agreement with the reference spectrum. In particular, the Cf-252 unfolded data shown in fig. 5 shows very good agreement with the reference spectrum in the entire energy range. The plutonium-oxide data shown in fig. 6 shows some oscillations in the energy range between 2.5 and 4.5 MeV. A possible reason for this disagreement is the lack of well-converged pulse-height distribution at these higher energies due to statistical fluctuations and the effect of the limited vertical range of the data acquisition electronics. The results presented show that the two sources can be discriminated from each other, a capability that is of interest to nuclear safeguards and nonproliferation application.

6. – Conclusions

The results presented in this paper show the promising capabilities and future potential of the SLS method for neutron source identification by unfolding pulse height distributions measured by liquid-scintillation detectors. We showed that SLS method can be used to efficiently unfold unknown spectra from measured neutron sources and bulk fissile material. We showed that the Cf-252 and plutonium oxide sample can be distinguished from each other within minutes, which is of paramount importance in nonproliferation applications. Future work will include an extensive analysis and assessment of the influence of statistical fluctuations, measurement time, measurement error, and shielding effects on the evaluated spectra data. The potential for source identification will also be evaluated.

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