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# Comparison of unfolding approaches for monoenergetic and continuous fast-neutron energy spectra

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## HIGHLIGHTS

- Proton-recoil detectors offer the possibility to unfold fast-neutron energy spectra.
- Pulse height distributions simulated by the MCNP-Polimi code.
- Maximum-Likelihood Expectation Maximization (MLEM), one-step-late (OSL) methods.
- Unfolding of monoenergetic and continuous-in-energy neutron sources.
- Study has shown the OSL method has superior unfolding performance to the MLEM method.

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## ABSTRACT

Proton-recoil detectors offer the possibility to unfold fast-neutron energy spectra of various sources. However, quantifying the confidence of the unfolding methodology is a complex task. In this paper, we present a comparative analysis of the maximum-likelihood, expectation-maximization (MLEM) method and one-step-late (OSL) method for neutron energy spectra unfolding. The analysis is performed on Monte Carlo simulated data for several monoenergetic neutron sources and continuous-in-energy $^{252}$Cf, $^{241}$Am–Be and $^{241}$Am–Li neutron sources. The results obtained for the monoenergetic neutron spectra show that both unfolding methods provide results that are in good agreement with the reference data. Very good agreement between the unfolded and the reference data is achieved for $^{252}$Cf, $^{241}$Am–Be, and $^{241}$Am–Li neutron spectra by using the OSL method. In the paper it is demonstrated that the MLEM and OSL methods can be applied to accurately unfold the simulated pulse-height distributions for organic liquid scintillation detectors. Comparative analysis between the two unfolding methods has shown that the OSL method has superior unfolding performance than the MLEM method.



## 1. Introduction

Investigation on various approaches to obtain accurate information on neutron energy spectra is of high interest in the fields of nuclear nonproliferation and homeland security, international safeguards, nuclear material control and accountability, national security, and counter-terrorism programs (Mullens et al., 2004). The neutron spectrum is a unique characteristic of each neutron source and can be used for relatively accurate source identification and characterization. For example, the assays performed on nuclear materials with organic scintillators, with a common objective of determining the mass and composition of the fissile samples, require accurate spectrum unfolding. Also, detailed knowledge of the fast-neutron spectrum is needed for accurate determination of neutron equivalent dose (Marinkovic et al., 1992).

Liquid and plastic organic scintillators are currently being investigated as the main components of various measurement systems. Liquid organic scintillators such as NE-213, EJ-301, and BC-501A are frequently used because of their good pulse-height response and their capability to accurately discriminate neutrons from γ-rays. Pulse-height spectra (PHS) obtained from such detectors are in fact proton-recoil spectra. Therefore, the neutron energy spectrum of the incident radiation has to be unfolded from a measured PHS.

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Techniques for neutron spectrometry by means of the spectrum unfolding have advanced considerably over the years. In the past, FERDOR (Burrus and Drischler, 1965) and FORIST (Johnson and Wehring, 1976) codes were among the first ones used for this purpose. A matrix inversion method to calculate a lower and upper bound for each element of the solution vector was a basis for these codes. Several other approaches to unfold PHS were developed; for instance, the maximum-entropy deconvolution code MAXED (Reginatto et al., 2002; Reginatto and Goldhagen, 1999) that uses a measured response matrix and "flat" default spectra. Neutron spectrum unfolding tools using a genetic algorithm (Mukherjee, 1999), a singular value decomposition (SVD) algorithm (Höcker and Kartvelishvili, 1996) a sequential least-squares algorithm (Xu et al., 2007) and artificial neural networks were developed more recently (Koohi-Fayegh et al., 1993; Avdic et al., 2006, Avdic et al., 2010). The Bayesian method is used to provide a formal foundation for the use of *a priori* information (Geman and Geman, 1984). The *a priori* information is not limited to data values and uncertainties but can include correlations and various physical constraints. Sometimes, the only *a priori* information might be that all the fluence values should be positive.

This paper presents a comparison of the maximum-likelihood, expectation-maximization (MLEM) method (Dempster et al., 1977) and one-step-late (OSL) method (Green, 1990) for unfolding of the simulated pulse-height distributions from liquid scintillation detectors. The simulated data for a few monoenergetic neutron sources, and continuous-in-energy $^{252}$Cf, $^{241}$Am–Be, and $^{241}$Am–Li neutron sources were obtained with the MCNP-PoliMi Monte Carlo code version 1.0 (Pozzi et al., 2003) and a specialized post-processing algorithm.

## 2. The simulated PHS

The PHS used in the unfolding procedures with the MLEM and OSL methods were obtained with the MCNP-PoliMi code and its post-processing algorithm. In this way, a simulated response for a liquid scintillator is obtained. In the MCNP-PoliMi code, each neutron interaction is accurately modeled, taking into account individual neutron energy depositions on the scintillator constituents. Specifically, elastic and inelastic scatterings on hydrogen and carbon nuclei are considered. The light output from secondary 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 final post-processing of the MCNP-PoliMi output is performed by taking into account the cumulative effect of multiple scatterings on the final light output.

The MCNP-PoliMi code was used to simulate the detector response matrix. Neutrons from a uniformly distributed surface source were emitted perpendicularly to the base of a 5 cm × 5 cm NE-213 cylinder with a H:C ratio of 1.21 and a density of 0.96 g/cm$^3$. The MCNP-PoliMi model of the detector is shown in Fig. 1. The relation between pulse height and neutron energy is described by the detector response matrix. Pulse height (light output) is usually expressed in MeVee units. A pulse height of E MeV electron equivalent (MeVee) is related to a signal with the same pulse height as that generated by an electron of energy E MeV. A part of the detector response matrix, simulated with the MCNP-PoliMi code, is shown in Fig. 2. In order to better visualize the response matrix, we selected neutron energies of 2, 5 and 8 MeV, 11 and 14 MeV in different pulse height ranges and different count rate ranges.

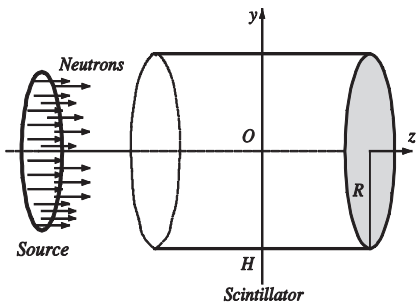


**Fig. 1.** The MCNP-PoliMi detector model.

The input PHS for the unfolding of monoenergetic (4, 8, and 12 MeV) and continuous energy spectra ($^{252}$Cf, $^{241}$Am–Be, and $^{241}$Am–Li) were acquired with the MCNP-PoliMi code, see Fig. 3.

## 3. Unfolding of the simulated PHS

The spectrum unfolding is the process of obtaining the energy distribution of the neutrons from the measured PHS. This 'inversed' procedure is ill-posed: small fluctuations of measured pulse heights can cause large uncertainties in the unfolded neutron-energy spectra. The inverse problem can be expressed in the following discrete form:

$$y_i = \sum_{j}^{J} r_{ij} x_j, \quad i = 1, \ldots, I \tag{1}$$

where $y_i$ is the binned number of counts 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.

### 3.1. Unfolding with the MLEM method

Maximum likelihood (ML) method is a popular statistical method used to make inferences about parameters of the underlying probability distribution of a given data set. The ML method is based on finding the value of one or more parameters for a given statistics which makes the known likelihood distribution a minimum.

The advantages of the ML methods are:

1. The methods provide a consistent approach to parameter estimation problems. This means that ML estimates can be developed for a large variety of estimation situations.
2. ML methods have desirable mathematical and optimizability properties. They become minimum-variance unbiased estimators as the sample size increases.

The disadvantages of the ML methods are:

1. The likelihood equations need to be specifically worked out for given distribution and estimation problem.
2. The numerical estimation is usually non-trivial.
3. ML methods can be sensitive to the choice of starting values.
4. ML estimates can be heavily biased for small samples. PHS are severely affected by Poisson noise, which implies that low data count rates give a less accurate prediction of number of neutrons received in an energy group.

A possible way to model the Poisson nature of the measures is to treat the data as stochastic variables instead of the exact values. When unfolding a Poisson distribution, without making any *a priori* assumptions about neutron spectrum distribution, the likelihood function equals to

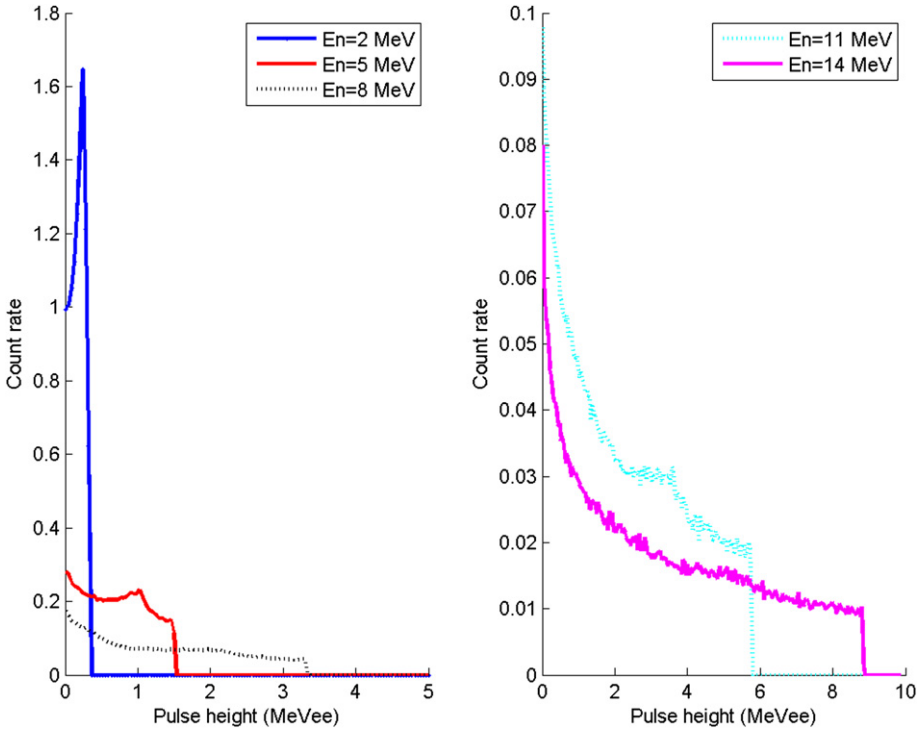


**Fig. 2.** A part of simulated detector response matrix for a NE-213 scintillator for the selected neutron energies. The matrix provides information on the conversion between the neutron energy and the corresponding pulse height (light output).

$$P_r = \prod_{i=1}^{I} \frac{\lambda_i^{y_i} e^{-\lambda_i}}{y_i!}, \tag{2}$$

where $\lambda_i = \sum_{j=1}^{J} r_{ij} x_j$ are the mean values of binned count rates $y_i$. The log-likelihood function then equals to

$$\ln P_r = \sum_{i=1}^{I} \left[ -\sum_{j=1}^{J} r_{ij} x_j + y_i \ln \sum_{j=1}^{J} r_{ij} x_j - \ln(y_i!) \right]. \tag{3}$$

Consequently, the $j$th component of the score equals to

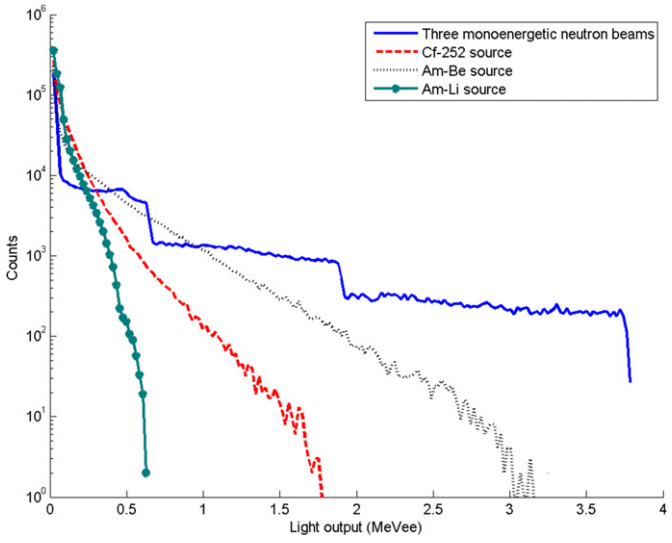


**Fig. 3.** The MCNP-PoliMi pulse-height spectra.

$$\frac{\partial \ln P_r}{\partial x_j} = \sum_{i=1}^{I} \left[ -r_{ij} + \frac{\lambda_i r_{ij}}{\sum_{l=1}^{J} r_{il} x_l} \right] = 0, \quad j = 1, \ldots, J \tag{4}$$

The algorithm to find nonnegative values of each $x_j$, such that all of the partial derivatives vanish is called the ML expectation maximization (MLEM as introduced earlier) algorithm. The $x_j$ values are calculated as follows:

$$x_j^{(k+1)} = \frac{x_j^{(k)}}{\sum_{i=1}^{I} r_{ij}} \sum_{i=1}^{I} r_{ij} \frac{y_i}{\sum_{l=1}^{J} r_{il} x_l^{(k)}}, \quad j = 1, \ldots, J \tag{5}$$

where $x_j^{(k)}$ represents the $k$th spectrum estimate. The term $\sum_{i=1}^{I} r_{ij}$ is the efficiency of the detector in the energy bin $j$.

The MLEM algorithm has the following important properties:

1. The algorithm increases the likelihood that the neutron spectrum estimate will generate the measured data until an absolute maximum during each iteration. However, as the number of iterations increases it also will enhance the noise level of the reconstructed data. Therefore, a balance between accuracy and variance has to be found.
2. Neutron spectrum is constrained to remain positive in each iteration.
3. The algorithm takes into account the Poisson nature of the noise in the PHS. This feature of the MLEM algorithm leads to the neutron spectra that are not as noisy as, for example, spectra obtained by means of least square method.
4. The drawback of the MLEM algorithm is that the neutron spectrum reconstruction is slow (large number of iterations is required), especially when accurate (i.e. large) response matrices are used.

### 3.2. Unfolding by the OSL method

A common problem with MLEM is that it generates noisy neutron energy spectra as the iterations proceed. In order to avoid this problem in the OSL algorithm, the iterations should be stopped before the convergence. This approach, however, suffers from a noise-bias tradeoff. If the convergence is reached, the solution is too noisy. On the other hand, if a small number of iterations is used, the solution is less noisy, but the quantitative level of solution values are biased towards the initial solution. This indicates it is not straightforward to estimate the unfolding error.

Empirical evidence suggests that OSL algorithm usually converges for properly chosen $\beta$. The deterioration in neutron spectrum quality can be measured by normalized mean square error (MSE) between the simulated or measured proton spectrum data and the neutron spectrum as a function of iteration index. The minimum of MSE can be used as the stopping criteria for OSL.

The concept of changing an ill-posed reconstruction problem into a well-posed one is based on introducing an extra control on what solutions are more favorable than the others. A 'good' solution is required not only to match the data as closely as possible, but also to be consistent with additional criteria that are set independently of the data. The objective function to be maximized is not the likelihood (Eq. (3)) but *a posteriori* probability density function. The prior is described by the Gibbs distribution $P(x) \propto \exp(-\beta U(x))$ (Geman and Geman, 1984), the penalty term, where $\beta$ is the scalar weighting parameter, and $U(x)$ is the energy function.

One of the Bayesian formulations, the OSL algorithm, uses a current estimate $x^{(k)}$when calculating the value of the derivative of the energy function $U$ (Green, 1990). Next iteration can be calculated as

$$x_j^{(k+1)} = \frac{x_j^{(k)}}{\sum_{i=1}^{I} r_{ij} + \beta \frac{\partial U(x,j)}{\partial x_j}} \sum_{i=1}^{I} r_{ij} \frac{y_i}{\sum_{l=1}^{J} r_{il} x_l^{(k)}}, \quad j = 1,\ldots,J \tag{6}$$

where $U(x,j)$ is the energy function defined as

$$U(x,j) = \sum_{b \in N_j} w_{jb} v(x_j - x_b) \tag{7}$$

where $w_{jb}$ is the weight of energy bin $b$ in the neighborhood of energy bin $j$ and the parameter $\beta$ is the confidence of the prior. In our investigation, we used a well-known choice of quadratic prior $v(r) = r^2$. The advantage of the quadratic energy functions is that their derivatives are linear. This makes it very simple to solve the maximization problem and to achieve high performance of the MLEM algorithm.

The regularization parameter $\beta$ controls the degree to which the models of the prior are accentuated, and keeps constant during the iterations. We must empirically select an appropriate value for the parameter $\beta$. Parameter $\beta$ is roughly estimated by means of greedy strategy. Plot of energy function for final solution after some prescribed number of iterations at parameter $\beta$ versus $\beta$ and the minima of the curve, should determinate the optimum value of $\beta$. This is because energy function can be interpreted as a measure of the total roughness of the neutron spectrum. For the low value of $\beta$, the solution is dominated by likelihood function and this implies that the roughness in the solution tends to rise for noisy data. For the high value of $\beta$, the prior dominates in the solution and the parasite effect of oversmoothing may take place. Although energy function may be small in the preceding iteration, a high value of $\beta$ excessively emphasizes the roughness measured by energy function, and in a current iteration, the spectrum may be less smooth.

If $\beta = 0$, Eq. (7) is reduced to the usual MLEM algorithm. In Eq. (7), one can clearly see that the prior is computed using the solution vector obtained in the previous iteration.

The numerical implementation of the iterative algorithms (both MLEM and OSL) includes the initial guesses of the estimated neutron fluences. In all calculations we have used the same positive values for starting guesses of neutron fluences in all energy groups. The results of preliminary analysis have shown that the iterative procedures applied for estimation of neutron energy spectra virtually do not depend on the choice of starting values in a relatively wide range of initial values.

## 4. Unfolding results

The MLEM and OSL unfolding methods require accurate knowledge of the detector response matrix. In the response matrix, each row corresponds to a neutron energy and each column corresponds to a pulse height. The MCNP-PoliMi code was used to calculate the NE-213 detector response matrix with PHS simulated in 256 channels and neutron energy range up to 15 MeV with an energy step of 0.1 MeV. For the evaluation of $^{241}$Am–Li neutron spectra we used the detector response matrix in a shorter energy range up to 4 MeV in order to evaluate the low-energy $^{241}$Am–Li spectrum with higher accuracy.

### 4.1. Monoenergetic neutron sources

Fig. 4 shows the results obtained by using the MLEM and OSL unfolding methods for three monoenergetic sources. We investigated neutron peaks of equal probability with energies equal to 4, 8, and 12 MeV. It can be seen that the MLEM method correctly predicts energy of sharply unfolded neutron peaks with energy resolution of approximately 0.1 MeV, but the intensities of the peaks are not equal. The peak at 4 MeV matches the reference data, but the peaks at 8 MeV and 12 MeV underestimate the references by 15 and 5%, respectively.

The OSL method identifies the three peaks correctly. The peak at 4 MeV is correctly estimated, but the peaks at 8 and 12 MeV underestimate the references by 20 and 15%, respectively. It can be

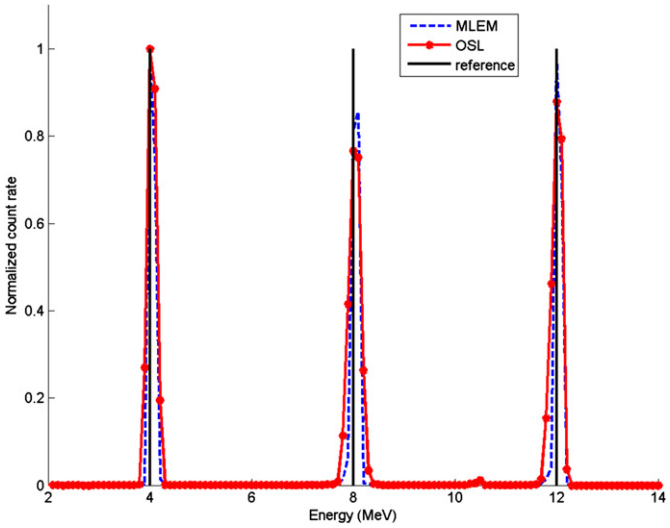


**Fig. 4.** Unfolding results for three monoenergetic neutron beams. The energies are correctly estimated with both methods.

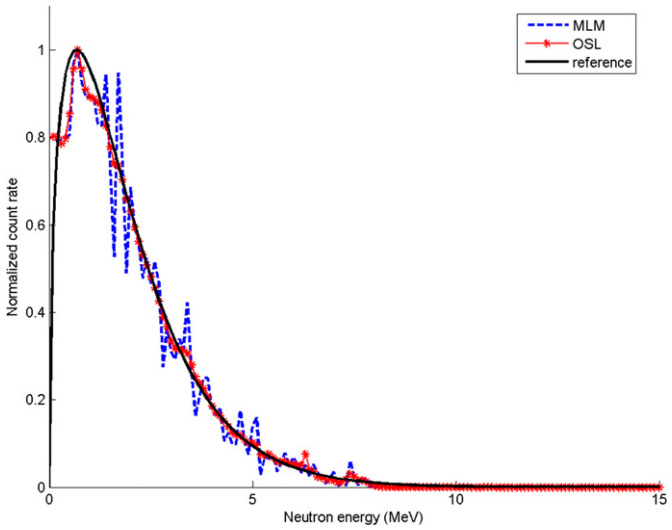


**Fig. 5.** Unfolded spectra of the $^{252}$Cf neutron source from the simulated data. Number of iterations was set to 1000 for both cases. The OSL unfolding was performed using $\beta = 5{\cdot}10^{-4}$.

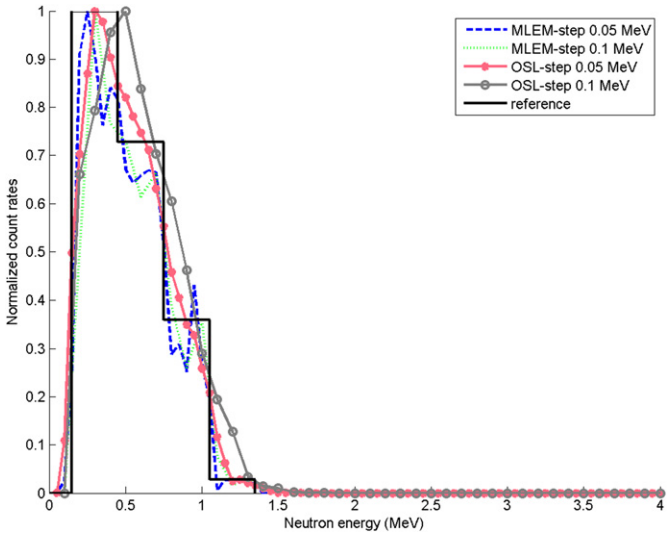


**Fig. 7.** Unfolded spectra of the $^{241}$Am–Li neutron source from the simulated data. Number of iterations was set to 1000 for both cases. The OSL unfolding was performed using $\beta = 5{\cdot}10^{-4}$.

noticed that the peaks obtained by the OSL method are slightly wider compared to the results obtained with the MLEM method, by up to 25%. This effect is expected because the regularization incorporated in the OSL method to reduce the oscillations of the spectra contributes to the width of the peaks.

### 4.2. Continuous-in-energy neutron sources

The MLEM and OSL methods were applied to continuous neutron energy spectra for the $^{252}$Cf, $^{241}$Am–Be, and $^{241}$Am–Li neutron sources.

The results obtained by applying the MLEM and OSL methods to the simulated PHS for the $^{252}$Cf source are shown in Fig. 5. The results are normalized to the maximal value of the spectra. Both methods provide unfolded spectra that are in relatively good agreement with the shape of the reference spectrum. The result obtained with the OSL method is smoother than that obtained with the MLEM method and generally in better agreement with the reference data.

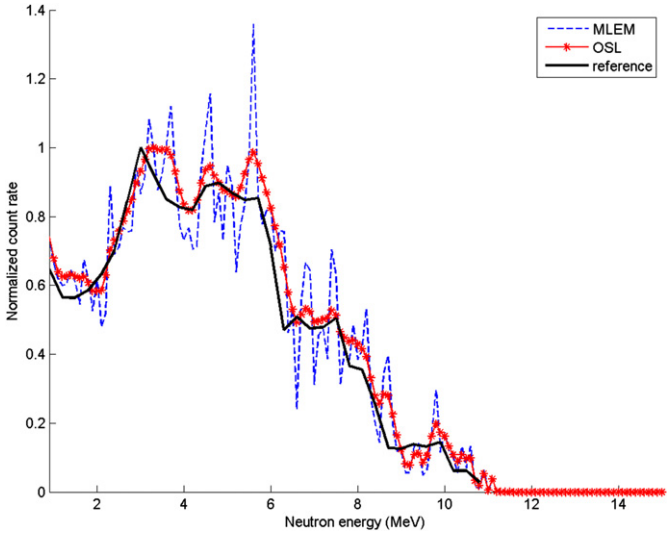


**Fig. 6.** Unfolded spectra of the $^{241}$Am–Be neutron source from the simulated data. Number of iterations was set to 1000 for both cases. The OSL unfolding was performed using $\beta = 5{\cdot}10^{-4}$.

The results obtained by the MLEM and OSL methods for the $^{241}$Am–Be source are presented in Fig. 6. The OSL results and the reference spectrum were normalized to the maximal value of the peak at 3 MeV. The result for the OSL follows the shape of spectrum whereas the MLEM result shows significant oscillations, although it does generally follow the trend of the reference data.

Fig. 7 shows the unfolded spectra of the $^{241}$Am–Li neutron source from the simulated data with the same values of the beta parameters as for $^{252}$Cf and $^{241}$Am–Be sources. The results of this study have shown that it is not possible to evaluate the $^{241}$Am–Li spectrum with good accuracy by using the response matrix in the neutron energy range up to 15 MeV, since the reference $^{241}$Am–Li spectrum is given in very few points in the range between 0 and approximately 1.5 MeV. In order to overcome this obstacle with few spectral data and a wide energy range, we have simulated the matrix in a shorter energy range with the MCNP-PoliMi code.

We have used two response matrices in the same neutron energy range up to 4 MeV but with two different energy bins of 0.1 and 0.05 MeV. It can be noticed in Fig. 7 that the OSL unfolding results with the response matrix with the smaller energy step show somewhat better agreement with the reference spectrum (Padovani et al., 2006). Since the $^{241}$Am–Li spectrum is the low-energy spectrum, the energy width of the channels should decrease taking into account that counting statistics is better for lower energy range than for higher neutron energies.

The results obtained, especially by the OSL method, have shown that it is possible to achieve the good agreement with the reference data (Padovani et al., 2006), by using the simulated detector response matrix with finer energy binning of 0.05 MeV in the energy range up to 4 MeV.

## 5. Conclusion

The presented unfolding results show the capabilities of the MLEM and OSL methods for the unfolding of PHS obtained with the MCNP-PoliMi simulation code. It was demonstrated that both methods can be used to accurately unfold unknown spectra from the simulated monoenergetic and continuous neutron sources. Analysis of the results obtained in this study has shown that the

OSL method provides more accurate results when compared to the MLEM method, especially for continuous neutron energy spectra. For the unfolding of monoenergetic sources, the MLEM method provides sharper peaks with better agreement with the reference intensities since the regularization incorporated in the OSL method contributes to the final width of the peaks. In the future, performances of the MLEM and OSL methods will be analyzed for experimental data from various neutron sources.

## Acknowledgements

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