# A STUDY OF POSSIBILITY TO DESIGN A FAST NEUTRON SPECTROMETER BASED ON THE ORGANIC SCINTILLATOR WITH SURROUNDING MATERIALS

by

***Senada AVDIĆ [1*], Predrag MARINKOVIĆ [2], Alma OSMANOVIĆ [1], Izet GAZDIĆ [1], Šejla HADŽIĆ [1], and Damir DEMIROVIĆ [3]***

[1]Department of Physics, Faculty of Science, University of Tuzla, Tuzla, Bosnia and Herzegovina
[2]Faculty of Electrical Engineering, University of Belgrade, Belgrade, Serbia
[3]Faculty of Electrical Engineering, University of Tuzla, Tuzla, Bosnia and Herzegovina

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**This paper deals with the design of a novel spectrometer of fast neutrons in nuclear safeguards applications based on the liquid organic scintillator EJ-309 with materials of different thickness surrounding the detector. The investigation was performed on the simulated data obtained by the MCNPX-PoliMi numerical code based on the Monte Carlo method. Among the various materials (polyethylene, iron, aluminum, and graphite) investigated as layers around the scintillator, polyethylene and iron have shown the most promising characteristics for evaluation of fast neutron energy spectra. The simulated pulse height distributions were summed up for each energy bin in the neutron energy range between 1 MeV and 15 MeV in order to obtain better counting statistics. The unfolded results for monoenergetic neutron sources obtained by a first order of Tikhonov regularization and non-linear neural network show very good agreement with the reference data while the evaluated spectra of neutron sources continuous in energy follow the trend of the reference spectra. The possible advantages of a novel spectrometer include a less number of input data for processing and a less sensitivity to the noise compared to the scintillation detector without surrounding materials.**

*Key words: neutron source, organic scintillator, shielding material, unfolding method, MCNPX_PoliMi code*

## INTRODUCTION

There is an interest of high importance in nuclear non-proliferation and nuclear safeguards applications to develop tools for robust and accurate identification of neutron sources. A proposed novel type of neutron spectrometer includes the liquid scintillator EJ-309 which is widely used in non-proliferation applications [1] and materials such as polyethylene (PE) or/and somewhat heavier materials of different thickness around the detector. The PE material is well-known by its property to slow down the neutrons. Neutrons lose more energy on collisions with light nuclei such as hydrogen compared to the heavy nuclei. The attenuation of fast incident neutrons becomes stronger with decreasing the neutron energy. The probability that an incident fast neutron reaches the detector will decrease as the moderator material is made thicker. On the other hand, the probability of absorption increases with increasing moderator thickness since absorption cross-sections have larger values at lower energies of neutrons [2]. It can be expected that the neutrons of higher energy will reach the detector with a higher probability compared to the neutrons of lower energy. The characteristics mentioned above provide a possibility to unfold the neutron energy spectra from the data acquired for various thickness of PE and/or other materials surrounding the liquid scintillation detector. However, for neutrons at higher energies (more than 10 MeV), the elastic interaction with the hydrogen is not effective in slowing down. The iron (Fe), as a material with good inelastic scattering properties, can cause a large change in neutron energy for high energy neutrons but have a little effect on neutrons at lower energy.

In order to design a new neutron spectrometer consisted of the scintillator detector with the surrounding materials, we have examined the possibilities of neutron spectra unfolding by using different methods. In addition to the traditional unfolding methods, it was demonstrated that the artificial neural network (ANN) [3] is a promising approach in the field of neutron spectrometry [4]. We constructed and trained the

* Corresponding author; e-mail: aadeno@bih.net.ba

feedforward backpropagation neural networks of different structure to unfold both monoenergetic and continuous neutron energy spectra in various energy ranges with different energy steps and PE and Fe layers of different thickness. In order to reduce the ambiguity in interpretation of the unfolded results we have also applied the least square method with a first order of Tikhonov regularization [5] to evaluate the energy spectra of monoenergetic neutrons and more complex spectra such as for $^{241}$AmBe and $^{252}$Cf spontaneous neutron fission source.

## THE MCNP-POLIMI SIMULATION OF THE NEUTRON SPECTROMETER WITH THE PE LAYERS

The MCNP-PoliMi code [6] has been used to model a new type of spectrometer with the scintillator and the PE or Fe enclosing materials. The spectrometer is consisted of a liquid scintillator of cylindrical shape with dimensions 13 cm  13 cm with a H:C ratio of 1.21 and a density of 0.935 g/cm$^3$ and PE or Fe blocks of various thickness with density of 0.955 g/cm$^3$ and of 7.87 g/cm$^3$, respectively.

First, we have investigated a possibility to design a spectrometer with a scintillation detector and the PE surrounding material. The MCNP-PoliMi simulation was performed for the EJ-309 detector, taking into account the PE layers placed in front of the detector in different ranges of thickness (between 1 cm and 10 cm, 1 cm and 15 cm, 1 cm and 26 cm) with the various thickness step of 0.5 cm, 1 cm, and 2 cm.

The MCNP-PoliMi code has been used for an accurate modelling of each neutron interaction in active volume of the detector as well as in PE blocks with thickness up to 26 cm. The MCNP-PoliMi code takes into account the individual neutron energy depositions through elastic and inelastic scattering on hydrogen and carbon nuclei as the main constituents of the scintillator material. The light output from the secondary particles produced by the neutron interactions within the scintillator is calculated on the basis of experimentally determined parameters [7]. The final light output was determined by the post-processing of the MCNP-PoliMi output considering the cumulative effect of multiple scatterings.

In all MCNP-PoliMi calculations we have simulated a parallel beam of neutrons as a monoenergetic neutron source in energy range between 1 and 15 MeV and various energy steps. For neutron sources with continuous energy spectra such as $^{252}$Cf and $^{241}$Am-Be we have used the same geometry of the incident neutron beam which was emitted perpendicularly to the PE blocks.

The detector response matrix represents connection between the neutron energy and the light output. The 3-D representation of the spectrometer response matrix for monoenergetic neutron sources between 1 and 15 MeV and 8 PE layers with thickness of 2 cm between 0 and 16 cm obtained by the MCNP-PoliMi code is shown in fig. 1. The simulated data relating the total number of counts per neutron for each energy in the range between 1 and 15 MeV with step of 0.1 MeV and 8 PE layers of different thickness are presented in fig. 2.

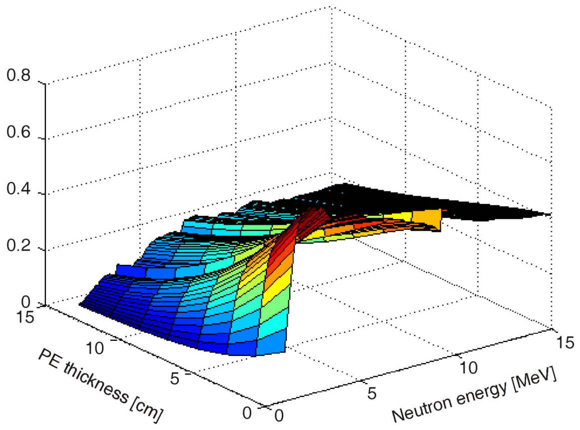


**Figure 1. The 3-D representation of the spectrometer response matrix for the monoenergetic neutron sources between 1 and 15 MeV with energy step of 0.1 MeV and 8 PE layers with thickness of 2 cm between 0 and 16 cm**

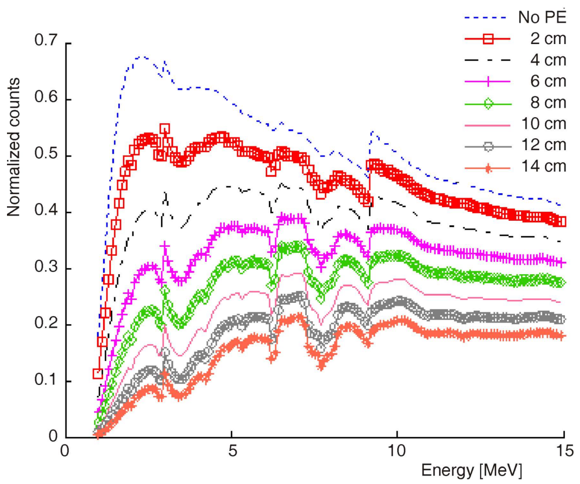


**Figure 2. The simulated data relating the total number of counts for each energy in the range between 1 MeV and 15 MeV with step of 0.1 MeV and 8 PE layers of different thickness**

We have investigated the iron (Fe) as a heavier material around the detector. Since for the neutrons at higher energies, the PE material is not effective in slowing down, we have selected Fe as a material with good inelastic scattering properties that can cause a large change in neutron energy higher than 10 MeV.

The 3-D representation of the spectrometer response matrix for monoenergetic neutron sources between 1 and 15 MeV with energy step of 0.5 MeV and Fe layers with thickness of 0.5 cm in the range between 0 and 20 cm obtained by the MCNP-PoliMi code is shown in fig. 3. The simulated data relating the total

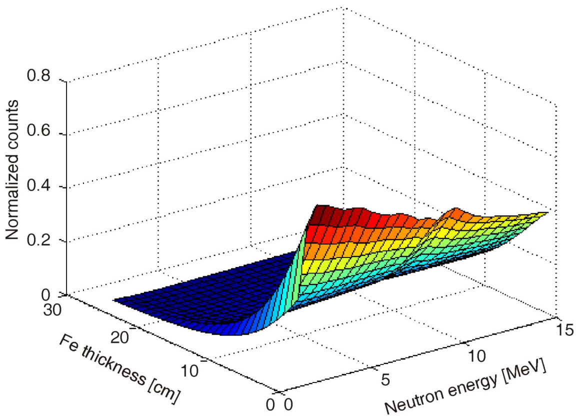


**Figure 3. The 3-D representation of the spectrometer response matrix for the monoenergetic neutron sources between 1 and 15 MeV with energy step of 0.5 MeV and 40 Fe layers with a step of 0.5 cm between 0 and 20 cm**

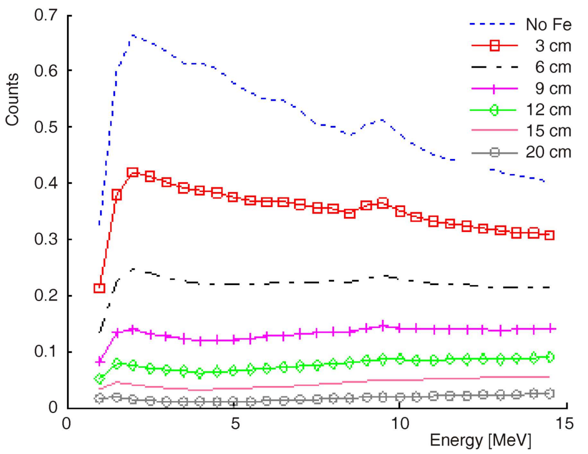


**Figure 4. The simulated data relating the total number of counts for each energy in the range between 1 MeV and 15 MeV with step of 0.5 MeV and 40 Fe layers of different thickness with a step of 0.5 cm**

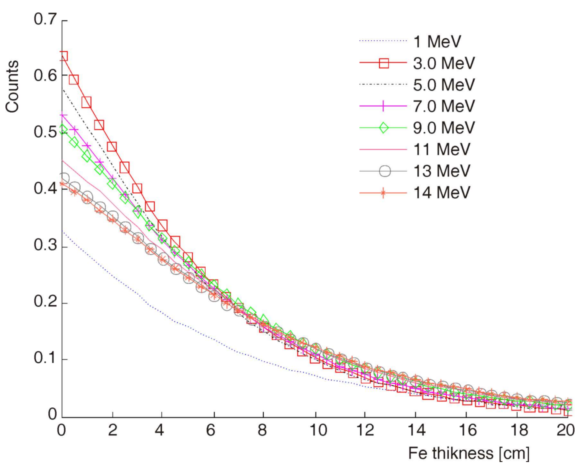


**Figure 5. The attenuation of neutrons with different energies through Fe layers of various thickness**

number of counts per neutron for each energy in the range between 1 and 15 MeV with step of 0.5 MeV and 40 Fe layers of different thickness are presented in fig. 4. The attenuation of neutrons of various energies in a Fe material has been shown in fig. 5.

## THE UNFOLDING RESULTS FOR THE DETECTOR WITH THE PE SURROUNDING MATERIAL

The unfolding problem can be described by the Fredholm integral equations of the first kind but discretization of such kind of integral can cause discrete ill-posed problems. The difficulty of solving the ill-posed problems lies in the fact that they are unstable and extremely sensitive to noise. When the problem $Ax = b$ is not well-posed (either because of non-existence or non-uniqueness of $x$), then the standard approach known as linear least squares must be modified. Some form of regularization is required in order to compute a stabilized meaningful approximate solution to discrete the ill-posed problems. It is necessary to incorporate further information about the desired solution in order to obtain a useful and stable solution [4].

In order to reduce the ambiguity in interpretation of the unfolded results we have applied the least square method with a first order of Tikhonov regularization and neural network approach.

### The neutron energy spectra evaluated by neural network approach

The ANN algorithms provide a very powerful alternative for the solution of the unfolding problem [8]. In order to unfold the neutron energy spectra from the simulated pulse height distributions data and their sum for each PE layer we have constructed a few non-linear neural networks for different energy resolutions. The networks were trained with the monoenergetic and continuous energy spectra ($^{251}$Cf and $^{241}$AmBe) in the energy range between 1 and 15 MeV with different energy steps of 0.1, 0.2, 0.4, and 0.7 MeV. The normalized number of counts for each thickness of PE layer up to 16 cm with step of 2 cm have been used as input data while the energy spectra of the incident neutrons with monoenergetic and continuous energy distributions have been used as output data.

The structure of the network has been determined on the basis of trail and error. After the trail and error procedure, the ANN sigmoid function was chosen as well as the number of hidden layers and the number of neurons in the hidden layers. For the energy step of 0.7 MeV we have designed the multilayer feedforward network with backpropagation algorithm with 22 energy groups as target data and the hidden layer consisting of 45 neurons while the number of input and output nodes corresponds to the number of PE layers and energy groups, respectively. Among various training algorithms available in Matlab Neural Network Toolbox [9], for this particular case with a small number of data, the Levenberg-Marquardt algorithm has shown the best performances. The neural network for energy step of 0.4 MeV consisted of one hidden layer with 55 neurons and 37 outputs with the Levenberg-Marquardt algorithm was used as a training algorithm. Good agreement between the unfolded results and the target data was achieved for both monoenergetic and continuous sources for the training data set. The input data set was too small to apply the complete the procedure for training, validation and testing of the network. For energy step of 0.1 MeV we have constructed the neural network with two hidden layers with 30 and 55 neurons and 142 outputs and resilient backpropagation algorithm was used as a training algorithm. In addition, a maximum likelihood method with OSL algorithm [10] for evaluation of monoenergetic neutron spectra has also been applied. The unfolded results obtained for the training data for both monoenergetic and continuous sources have not been in good enough agreement with the target data so that the better results could not be expected for the test data which are not presented to the network during the training procedure (fig. 6).

The energy step of 0.2 MeV was the smallest step for which we could apply the complete procedure of the network training including the validation and testing. We have constructed the neural network with two hidden layers with 45 and 25 neurons and 71 outputs. The network was trained with monoenergetic and sources with continuous energy spectra such as $^{252}$Cf and $^{241}$AmBe by using the resilient backpropagation training algorithm. The unfolded results shown in fig. 7 are in good agreement with the target data for a few randomly selected test monoenergetic neutron peaks never presented to the trained network before. The results shown in fig. 8. have been obtained for the trained data relating sources with the continuous energy distributions and they are in good agreement with the target data.

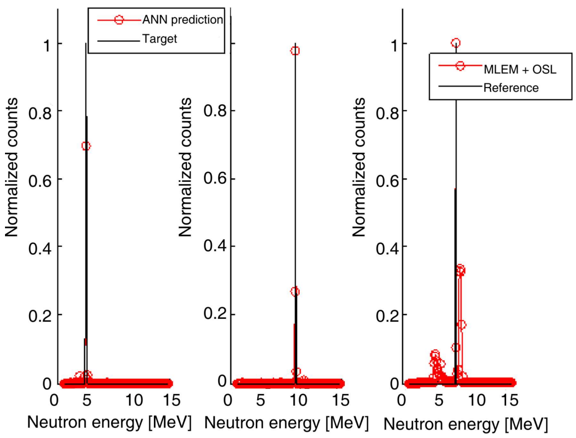


**Figure 6. The unfolded results for a few randomly selected monoenergetic neutron peaks with the energy step of 0.1 MeV obtained by the ANN approach and maximum likelihood with OSL algorithm**

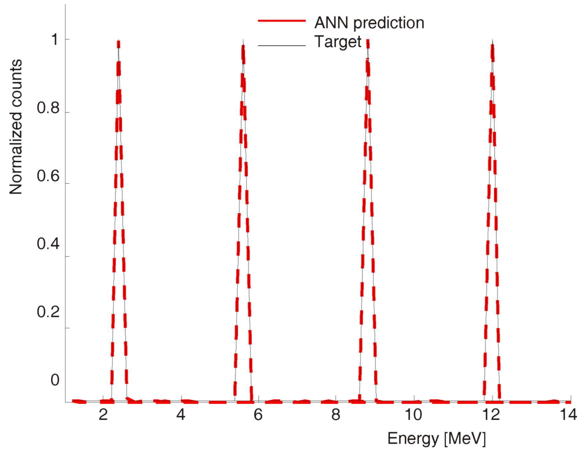


**Figure 7. The unfolded results compared with the target data for a few randomly selected test monoenergetic neutron peaks with energy step of 0.2 MeV never presented to the trained network before**

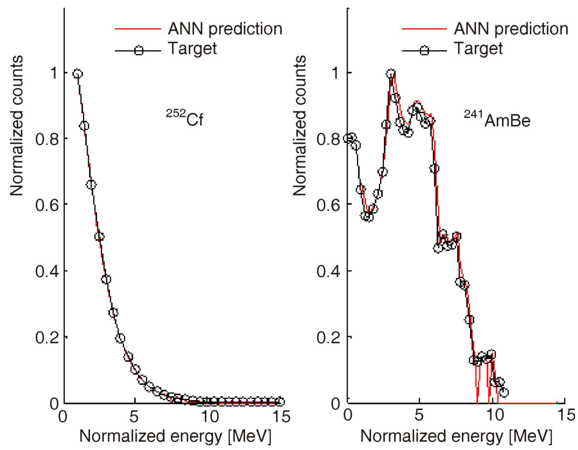


**Figure 8. The unfolded results for $^{252}$Cf and $^{241}$AmBe neutron sources obtained for the trained data with energy step of 0.2 MeV**

## The neutron energy spectra evaluated by the first order Tikhonov regularization

One of the most well-known forms of regularization in order to obtain a useful and stable solution of ill posed problem is the Tikhonov regularization [5]. If a system of linear equations $Ax = b$ is not well posed, then the standard approach is to minimize the residual $\|Ax \quad b\|$. The regularization term is included in this minimization in order to prefer a solution with desirable properties. The standard form of Tikhonov regularization is

$$\min\{\|Ax \quad b\|_2^2\} \quad \lambda\|Lx\|_2^2$$

where $\|Ax \quad b\|_2^2$ is the error term, $\lambda$ – the regularization parameter and $\lambda\|Lx\|_2^2$ – the penalty term. In zero order Tikhonov regularization, the regularization operator ($L$) is the identity matrix, while in the higher order Tikhonov, $L$ is either the first order or the second order differentiation matrix. We have used the first order of Tikhonov regularization. The regularization effect depends on $\lambda$ regularization parameter. For $\lambda = 0$, the regularization is reduced to the unregularized least squares problem.

In order to evaluate both monoenergetic and continuous energy spectra by the Tikhonov regularization we calculated the matrix for 31 PE layers of different thickness with step of 0.5 cm from 0 cm to 15 cm and 28 energy groups with energy step of 0.5 MeV in the range between 1 MeV and 15 MeV. The unfolded results for a few monoenergetic neutron peaks are in very good agreement with the target data (fig. 9). However, for the continuous energy distributions, the deviations between the unfolded and the target data are larger. A comparison of the unfolded spectrum by Tikhonov regularization and the reference $^{241}$AmBe spectrum is given in fig. 10.

With Tikhonov regularization we achieved a better resolution by encompassing the layers with step of 0.5 cm but worse energy resolution of 0.5 MeV compared to the results obtained by the nonlinear ANN with energy step of 0.2 MeV.

## THE UNFOLDING RESULTS FOR THE DETECTOR WITH Fe SURROUNDING MATERIAL

In order to investigate the iron as a surrounding material for a new spectrometer we have examined the effects of Fe layers on attenuation of monoenergetic neutrons and possibility to unfold the neutron energy spectra.

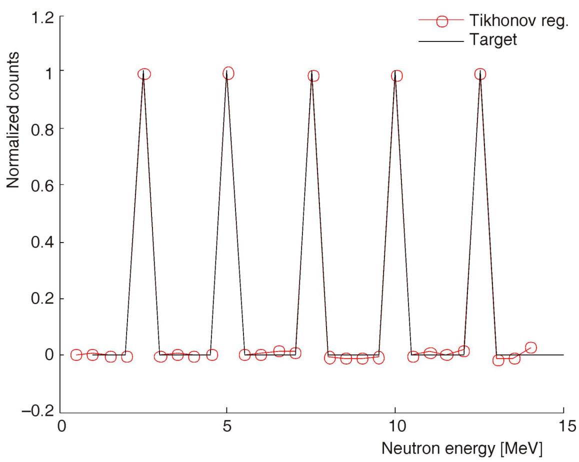


**Figure 9. The unfolded results obtained by the Tikhonov regularization for a few monoenergetic neutron peaks**

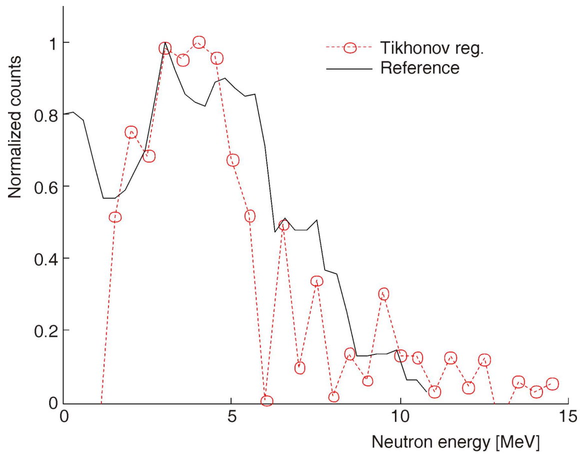


**Figure 10. The unfolded spectrum by Tikhonov regularization compared to the reference $^{241}$AmBe spectrum (without any type of smoothing)**

### The neutron energy spectra evaluated by the first order Tikhonov regularization method

Due to the deviations of the unfolded results with PE as a light material around the detector for sources continuous in energy from the reference ones, we have investigated Fe as a heavier surrounding material that can cause a large change in neutron energy for high energy neutrons.

The unfolding possibilities of a new spectrometer were investigated by the first order Tikhonov regularization method with the spectrometer response matrix for monoenergetic neutron sources between 1 MeV and 15 MeV with energy step of 0.5 MeV and 40 PE layers with a step of 0.5 cm between 0 and 20 cm.

It can be seen in fig. 11. that the unfolded results obtained by the Tikhonov regularization method for monoenergetic sources are in very good agreement with the reference data while figs. 12 and 13 for sources with continuous energy distributions show better agreement with the reference spectra compared to the results obtained for the spectrometer with PE as a material enclosing the scintillation detector.

For the energy step of 0.5 MeV in the range from 1 MeV to 15 MeV and the Fe step of 1 cm up to 26 cm we have calculated the response spectrometer matrix. The unfolded results for the monoenergetic and continuous sources have been shown in figs. 14 and 15, respectively. It can be seen that very good agreement between the monoenergetic neutron peaks and reference data was achieved by using the first order Tikhonov regularization, while for continuous sources such as $^{252}$Cf and $^{241}$AmBe, the deviations are somewhat larger compared to the previous results (given in figs. 12 and 13) but the energy spectra of these sources are still recognizable and follow the trend of the reference spectra.

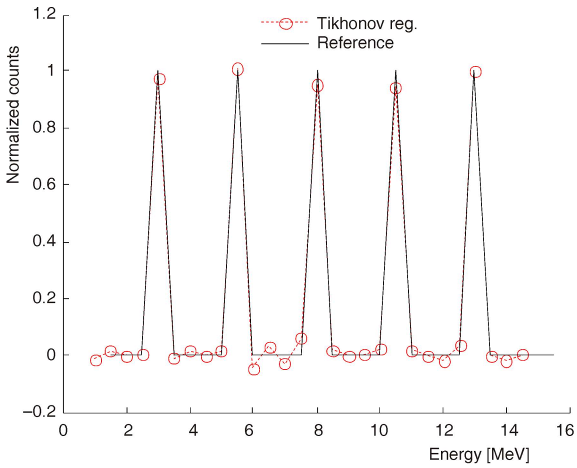


**Figure 11. The unfolded results for a few monoenergetic neutron peaks obtained by the Tikhonov regularization method compared to the reference data**

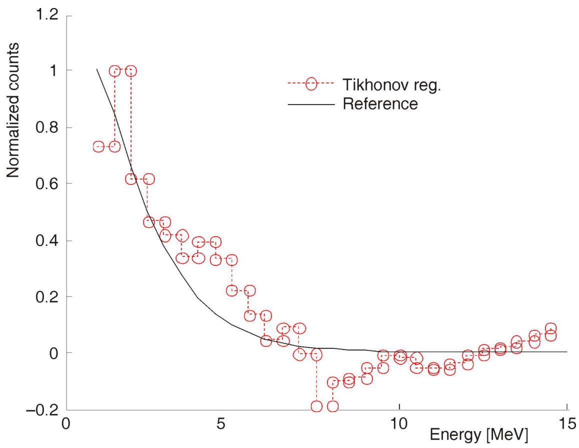


**Figure 12. The unfolded spectrum for $^{252}$Cf source obtained by the Tikhonov regularization method compared to the reference data**

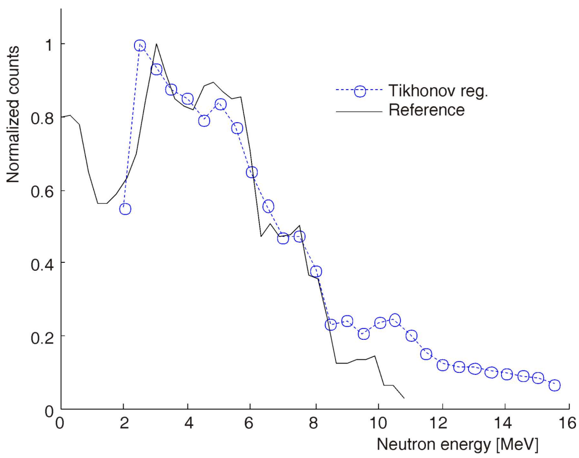


**Figure 13. The unfolded spectrum for $^{241}$AmBe source obtained by the Tikhonov regularization method compared to the reference spectrum**

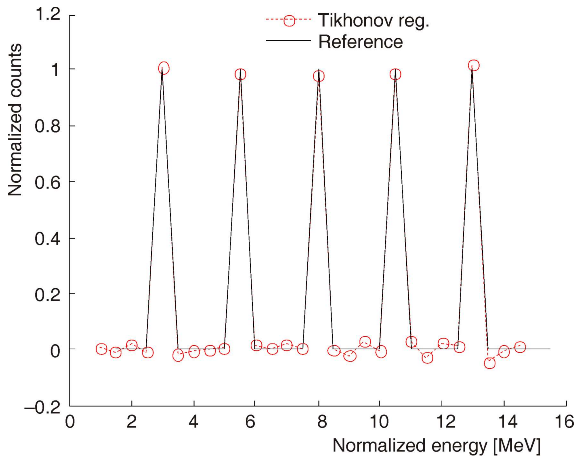


**Figure 14. The unfolded results for a few monoenergetic neutron peaks obtained by the Tikhonov regularization method compared to the reference data**

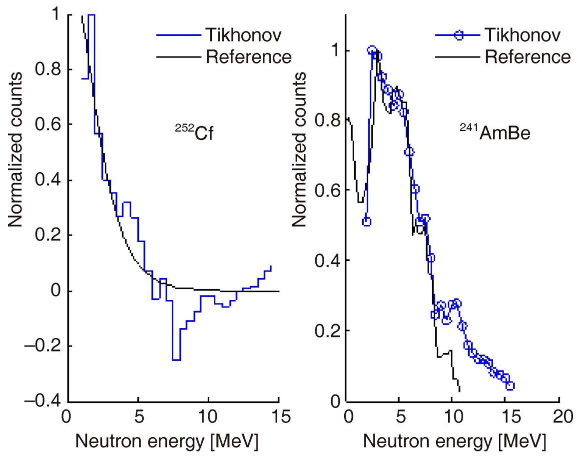


**Figure 15. The unfolded spectrum for $^{252}$Cf and $^{241}$AmBe sources obtained by the Tikhonov regularization method compared to the reference data**

## The neutron energy spectra evaluated by the neural network approach

Since the unfolded results obtained by the Tikhonov regularization method for the spectrometer with Fe as a surrounding material have shown that Fe can be potentially suitable for design of a novel spectrometer, we have constructed two different multilayer feedforward networks with backpropagation algorithm to further investigate the optimal step of enclosing layers. For an energy step of 0.5 MeV in the energy range from 1 MeV to 15 MeV, and a step of 1 cm up to 26 cm, we have designed the network with resilient training algorithm with 30 output nodes (28 energy groups for monoenergetic sources and 2 continuous sources such as $^{252}$Cf and $^{241}$AmBe). After the trial and error procedure we have chosen 2 hidden layers with sigmoid transfer functions consisting of 35 and 30 neurons, respectively, while the number of the input nodes corresponds to the number of Fe layers. The training procedure, including the validation and testing of the performances of the network, was applied. Among various training algorithms available in Matlab Neural Network Toolbox [10], for this particular case with small number of data, the Levenberg-Marqoard algorithm has shown the best performances. The unfolded results for the test data randomly selected among the monoenergetic sources that have not been shown to the network before are given in fig. 16. A comparison of the unfolded spectra for $^{252}$Cf and $^{241}$AmBe sources obtained by the ANN approach and the reference data has been shown in fig. 17.

It was demonstrated that it possible to obtain for a Fe matrix with 40 Fe layers and 28 energy groups with an energy step of 0.5 MeV from 1 MeV to 15 MeV and a Fe step of 0.5 cm up to 20 cm, the better agreement between the ANN prediction results and target data. We have designed the network with resilient training algorithm with 30 output nodes (28 energy groups for monoenergetic sources and 2 continuous sources such as $^{252}$Cf and $^{241}$AmBe) and 2 hidden layers with sigmoid transfer functions consisting of 38 and 33 neurons, respectively, while the number of input nodes corresponds to the number of Fe layers. The training, validation, and testing of the network were performed. The unfolded results for the test data randomly selected among the monoenergetic sources never presented to the network are given in fig. 18, while the spectra of the selected sources with the continuous energy distributions are presented in fig. 19.

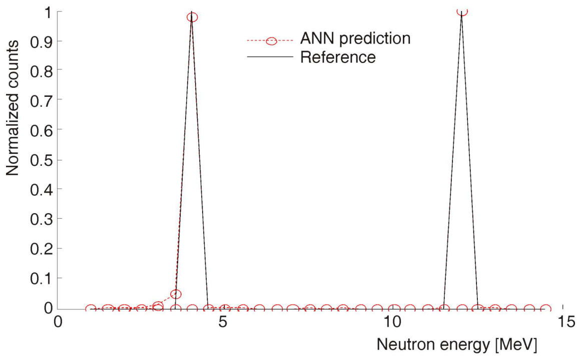


**Figure 16. The unfolded results for the test data with a Fe step of 1 cm that have not been presented to the network before**

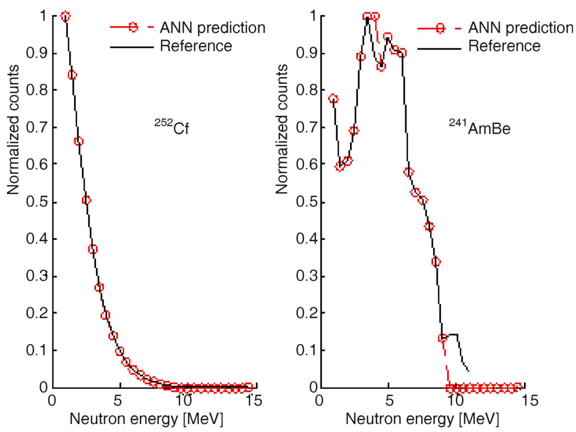


**Figure 17. The unfolded spectra for $^{252}$Cf and $^{241}$AmBe sources obtained by the ANN approach with a Fe step of 1 cm compared to the reference data**

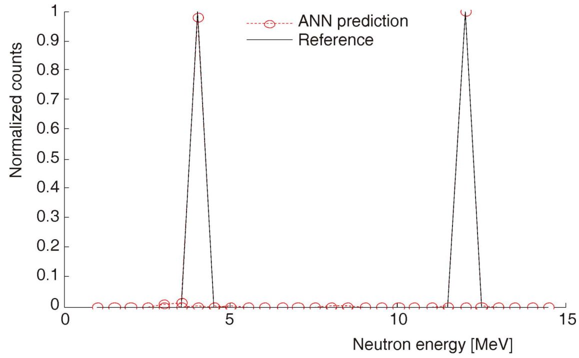


**Figure 18. The test data that have not been presented to the network before with a Fe step of 0.5 cm**

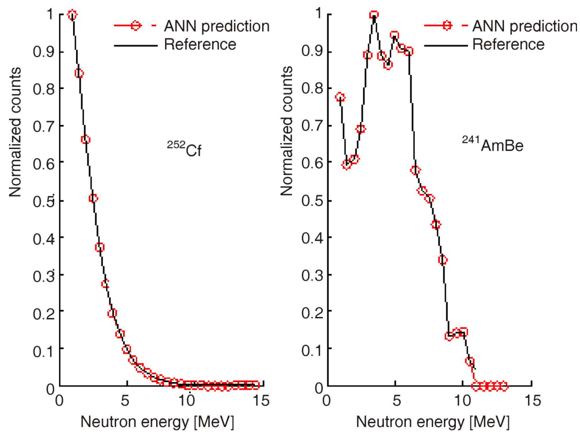


**Figure 19. The unfolded spectra of $^{252}$Cf and $^{241}$AmBe sources obtained by the ANN approach with a Fe step of 0.5 cm compared to the reference data**

The results of this study have shown that with a finer Fe step of 0.5 cm and a smaller range of Fe enclosing material up to 20 cm it was possible to obtain the unfolded results in better agreement with the reference data compared to the unfolded results obtained for the Fe step of 1 cm and the maximum thickness of Fe surrounding material up to 26 cm.

## CONCLUSIONS

Among the various materials (PE, Fe, Al, graphite) investigated as surrounding material around the EJ-309 liquid scintillation detector, the results obtained have shown that the iron has the most promising spectrometric characteristics since we could unfold the monoenergetic and continuous sources by using the first order of Tikhonov regularization and non-linear neural network as the two independent unfolding methods, with good agreement compared to the reference data. It was demonstrated that the polyethylene, as a light material, is suitable only for the evaluation of monoenergetic neutron spectra, while the materials such as graphite and aluminum do not have characteristics good enough for spectrometric purposes. For practical reasons, it is more appropriate to use a lighter material around the detector. It is predicted in the frame of the future work to investigate the spectrometric potential of a combination of thin layer of heavier material and wider layer of lighter material in order to meet the weight requirements for the spectrometer and to optimise the spectrometric performances.

Possible advantages of a novel spectrometer compared to the spectrometry by the EJ-309 without enclosing material are: much less number of input data to process and less sensitivity to the noise. In the next phase of research these possible advantages will be investigated experimentally.

## AUTHOR CONTRIBUTIONS

Numerical simulations were performed by S. Avdić, P. Marinković, A. Osmanović, I. Gazdić, and S. Hadžić. Software performance analysis tuning and optimization were carried out by D. Demirović. The manuscript was written by all authors.

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**Сенада АВДИЋ, Предраг МАРИНКОВИЋ, Алма ОСМАНОВИЋ, Изет ГАЗДИЋ , Шејла ХАЏИЋ, Дамир ДЕМИРОВИЋ**

## ИСПИТИВАЊЕ МОГУЋНОСТИ ПРОЈЕКТОВАЊА СПЕКТРОМЕТРА БРЗИХ НЕУТРОНА БАЗИРАНОГ НА ОРГАНСКОМ СЦИНТИЛАТОРУ СА ОКОЛНИМ МАТЕРИЈАЛИМА

Описан је дизајн новог спектрометра брзих неутрона за апликације у области нуклеарне сигурности. Дизајн је заснован је на течном органском сцинтилатору EJ-309 са материјалима различитих дебљина који окружују детектор. Испитивања су извршена са симулационим подацима који су добијени применом нумеричког кода MCNPX-PoliMi заснованог на Монте Карло методи. Између различитих материјала (полиетилен, гвожђе, алуминијум, графит) који су испитивани као слојеви око сцинтилатора, најперспективнија својства за евалуацију енергетских спектара брзих неутрона су показали полиетилен и гвожђе. Да би се добила боља бројачка статистика за сваки енергетски бин, амплитудска расподела импулса сумирана је за сваки енергетски корак у опсегу од 1 до 15 MeV. Резултати анфолдинга, добијени за моноенергетске неутронске изворе коришћењем Тихоновљеве регуларизационе методе првог реда и нелинеарне неуралне мреже, у веома доброј су сагласности са референтним подацима, док евалуирани спектри за неутронске изворе са континуалном енергетском расподелом следе ток референтних спектара. Добијени резултати су потврдили спектрометарски потенцијал који може бити побољшан комбинацијом различитих материјала око детектора. Могуће предности новог типа спектрометра укључују мањи број улазних података за обраду и мању осетљивост на шум у поређењу са сцинтилационим детектором без додатних слојева.

*Кључне речи: неутронски извор, органски сцинтилатор, окружујући материјал, анфолдинг метода, MCNPX_PoliMi код*