APLICATION OF WSP METHOD IN ANALYSIS OF ENVIRONMENTAL SAMPLES

***Matúš Stacho*[1]*, Štefan Krnáč*[2]*, Vladimír Slugeň*[1]*, Róbert Hinca*[1]*, Stanislav Sojak*[1]**

[1] *Institute of Nuclear and Physical Engineering, Faculty of Electrical Engineering and Information Technology, Slovak University of Technology,* [2] *METR, s.r.o.*
E-mail: matus.stacho@stuba.sk

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1. Introduction

Detection of activity in natural samples is specific especially because of its low level and high background interferences. Reduction of background interferences could be reached using low background chamber. Measurement geometry in shape of Marinelli beaker is commonly used according to low level of activity in natural samples.

The Peak Net Area (PNA) method is the world-wide accepted technique for analysis of gamma-ray spectra [1]. It is based on the net area calculation of the full energy peak, therefore, it takes into account only a fraction of measured gamma-ray spectrum. On the other hand, the Whole Spectrum Processing (WSP) approach to the gamma analysis makes possible to use entire information being in the spectrum [2]. This significantly raises efficiency and improves energy resolution of the analysis. A principal step for the WSP application is building up the suitable response operator. Problems are put in an appearance when suitable standard calibration sources are unavailable. It may be occurred in the case of large volume samples and/or in the analysis of high energy range. Combined experimental and mathematical calibration may be a suitable solution.

Many different detectors have been used to register the gamma ray and its energy. HPGe detectors produce the highest resolution commonly available today. Therefore they are they the most often used detectors in natural samples activity analysis. Scintillation detectors analysed using PNA method could be also used in simple cases, but for complicated spectra are practically inapplicable. WSP approach improves resolution of scintillation detectors and expands their applicability.

2. Whole spectrum processing

The whole spectrum processing (WSP) model is based on the response operator which is mathematically formulated by a vector model

$$d = K_c . q \tag{1}$$

where d is a column vector of the measured physical spectrum,
q is a column vector of the real incident spectrum and
K_c is a matrix of the complete response operator with dimension that corresponds to the length of physical and incident spectra [3].

As an aspect of statistical fluctuation in the gamma-ray spectra, a solution of (1) cannot be found by direct computation of the vector q (for example by direct inversion of K_c), and indirect iterative computational methods must be employed [3]. These methods are based on minimizing the residuum between physical and model spectra according to the vector q. The model fitting methods can be classified into two main groups:

a) the least squares (LS) approach, and
b) the maximum likelihood (ML) approach.

Then LS and ML residual functions may be expressed as:

$$\Delta_{LS} = (d - K_c.q)^2 \text{ and} \tag{2}$$

$$\Delta_{ML} = \log(d) - \log(K_c.q). \tag{3}$$

Using the residual function of LS or ML (or), the gradient method yields an iteration step for q that may by formulated as , where the symbol „grad“ represents derivates of the residual function according to all elements in the vector of q (gradient) and w is a length of the iteration step [3].

2.1. Response matrix operator K_c

Only few components of K_c matrix could be obtained by measured. Rest of the responses have to be supplemented to the matrix using Scaling Confirmatory Factor Analysis (SCFA) or by simulation of detector response. MCNP5 code was used to calculate detector responses to source in shape of Marinelli beaker. The energy range was from 10 up to 1750 keV. The responses were calculated for mono-energetic source for each keV in the energy range. Figures 1 and 2 depict used model of 2 x 2 inch NaI(Tl) detector with Marinelli beaker as volume source. The material composition is described in table 1.

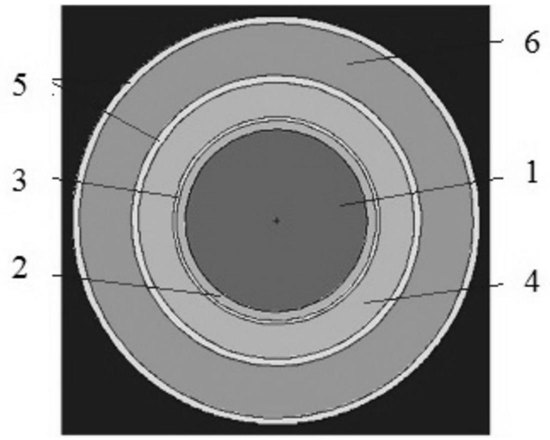

Fig. 1 *Axial section (1 - scintillator, 2 - reflector, 3 - casing, 4 - Surround, 5 – Marinelli beaker, 6 - source, 7 - vacuum)*

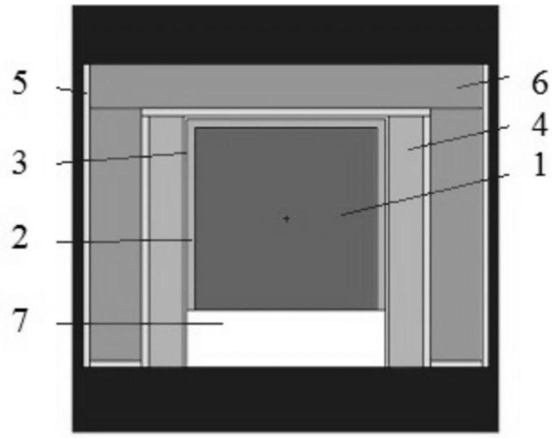

Fig. 2 *Vertical section (1 - scintillator, 2 - reflector, 3 - casing, 4 - Surround, 5 – Marinelli beaker, 6 - source, 7 - vacuum)*

Tab. 1 *Material composition of the model*

Part	Material	Modelled composition	Density [g.cm^{-3}]
Scintillator	NaI(Tl)	Na:I (1:1)	3.665
Reflector	MgO	Mg:O (1:1)	3.58
Casing	Aluminum	Al	2.7
Surround	Air	N:O:Ar (0,78:0,21:0,01)	0.0012
Marinelli beaker	Polyethylene	C:H (2:4)	0.9
Marinelli beaker content	Rubber	C:H (5:8)	0.98

Tally F8 (detector tally) was used to calculate detector response in active area. The calculation considers only interaction of the gamma rays with matter of detector. Peak energy broadening caused by flash collection and amplification in photo-multiplier tube was taken in to account using Gaussian Energy Broadening (GEB) function. GEB function is defined as:

$$FWHM = -0{,}01213 + 0{,}07329 * \sqrt{E - 0{,}2917E^2} \tag{4}$$

This dependence was obtained from calibration measurements. The complete response matrix operator K_c is shown in figure 4.

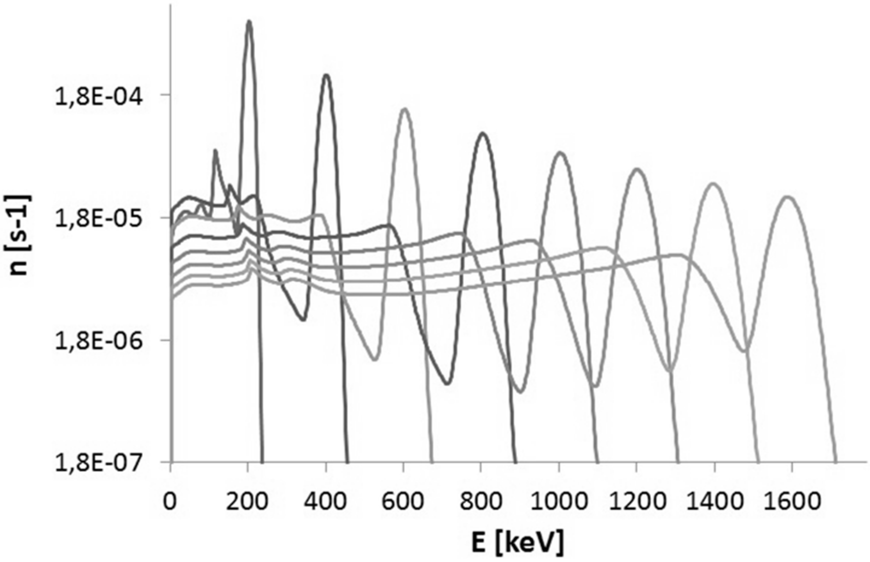

Fig. 3 *Response matrix operator* K_c.

3. Experimental

Matrix described in previous chapter was used to analyse few natural samples. The sample of dried mushrooms was chosen as an example. The sample was measured in the Marinelli geometry in low background chamber using 2 x 2 inch NaI(Tl) and 30% HPGe detectors. Figure 5 shows the spectrum collected by NaI(Tl) detector and the background spectrum.

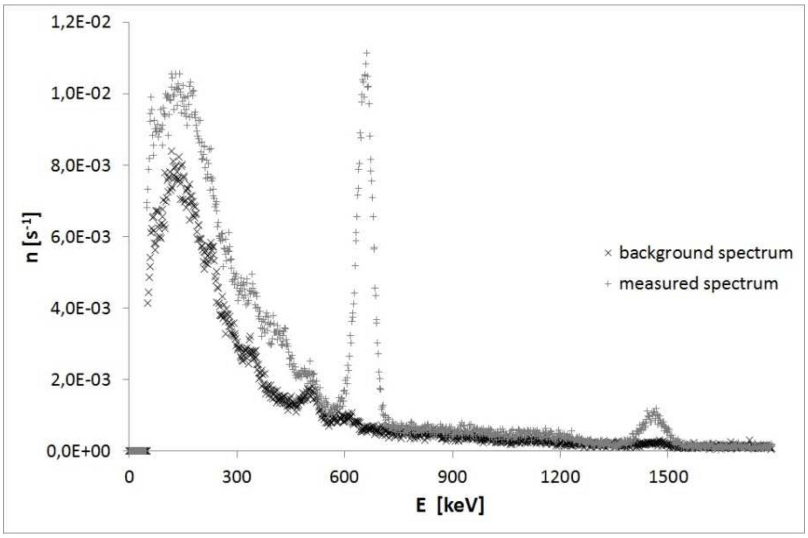

Fig. 4 *Measured and background spectrum*

Two dominant peaks in highest part of measured spectrum belong to ^{40}K and ^{137}Cs. The left low energy part contains also several peaks, but PNA method is inapplicable for analyze. The background spectrum was subtracted from the measured spectrum and analyzed using WSP method. Spectrum after subtraction of the background is shown in figure 6 as analyzed spectrum together with reconstructed spectra created during the analysis. The analysis result is together with the HPGe measurement shown in the figure 7. In the results could be seen significant improvement of the energetic resolution and separation of ^{137}Cs 661 keV peak from ^{214}Bi 609 keV peak. The lower part of the spectrum is still not reliably analyzable.

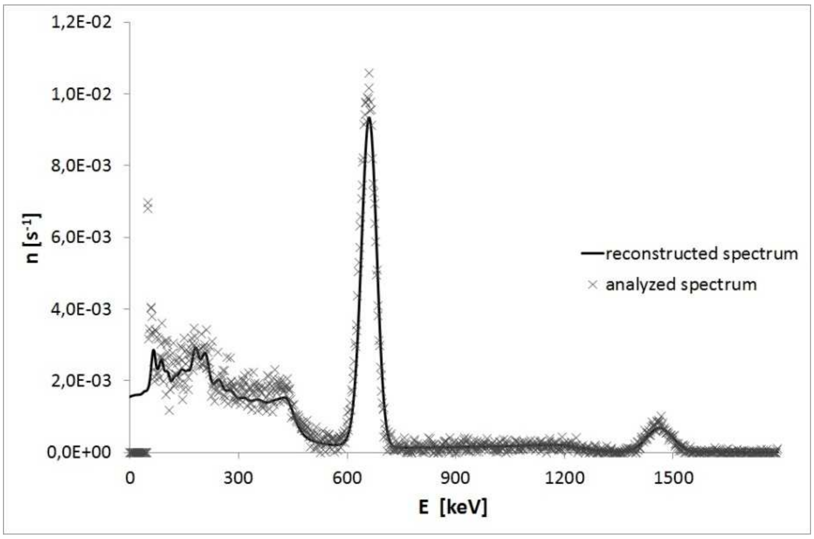

Fig. 5 *Analyzed and reconstructed spectrum*

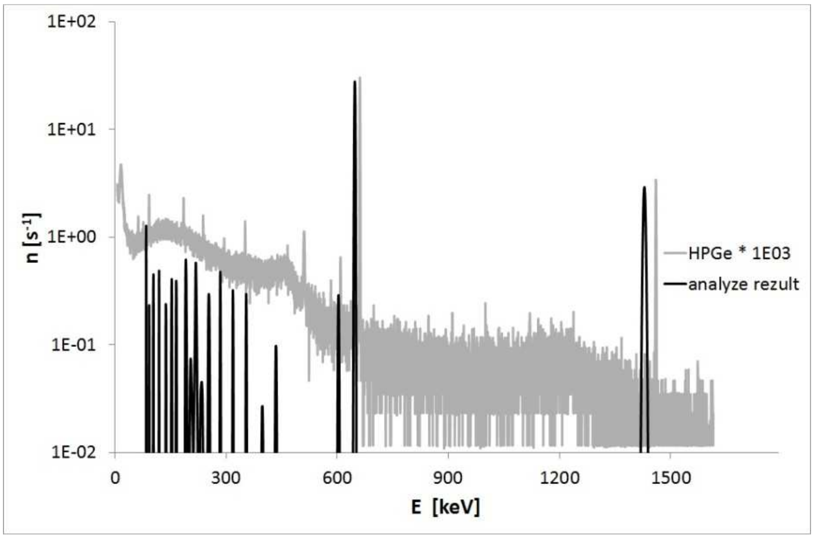

Fig. 6 *Analyze result and spectrum measured using HPGe detector*

4. Conclusion

WSP method allowed significant improvement of the energetic resolution and separation of ^{137}Cs 661 keV peak from ^{214}Bi 609 keV peak. At the other hand the statistical fluctuations in the lower part of the spectrum highlighted by background subtraction causes that this part is still not reliably analyzable.

Acknowledgement

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References:

[1] K. Debertin, R. G Helmer.: Gamma- and X-Ray Spectrometry with Semiconductor Detectors. Amsterdam_ North Holond (1998).

[2] Š. Krnáč, P. Povinec.*: Journal of Radioanalytical and Nuclear Chemistry*, 204 (1) 57, (1996).

[3] XXX Days of Radiation Protection., 10.-14.11.2008_Liptovský Ján_Slovak Republic_p. 103_(2008)