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Theoretical study of depth profiling with gamma- and X-ray spectrometry based on measurements of intensity ratios

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ABSTRACT

This article describes the method for the estimation of depth distribution of radionuclides in a material with gamma-ray spectrometry, and the identification of a layered structure of a material with X-ray fluorescence analysis. This method is based on the measurement of a ratio of two gamma or X-ray lines of a radionuclide or a chemical element, respectively. Its principle consists in different attenuation coefficient for these two lines in a measured material. The main aim of this investigation was to show how the detected ratio of these two lines depends on depth distribution of an analyte and mainly how this ratio depends on density and chemical composition of measured materials. Several different calculation arrangements were made and a lot of Monte Carlo simulation with the code MCNP – Monte Carlo N-Particle (Briesmeister, 2000) was performed to answer these questions. For X-ray spectrometry, the calculated Kα/Kβ diagrams were found to be almost independent upon matrix density and composition. Thanks to this phenomenon it would be possible to draw only one Kα/Kβ diagram for an element whose depth distribution is examined.

1. Introduction

The gamma- and X-ray spectrometry is a powerful tool for analysis of materials. Gamma spectrometry (GS) is used to identification and quantification of radionuclides, whereas the stimulated emission of X-ray radiation is applied to elemental analysis with an X-ray fluorescence (XRF) method. Samples for laboratory GS and destructive XRF are usually homogenized before the analysis. However, the homogeneous distributions of chemical elements or radionuclides are not ensured neither in non-destructive XRF nor in in-situ GS. The knowledge of depth distribution is essential information about an investigated object and it is also crucial for any quantitative analysis. The reasons for non-uniform distribution of radionuclides are natural phenomena in environment, i.e. surface contamination and migration of radionuclides (Ahmed, or Andersen et al., 2014, 2015), or it is caused by human activity (Trnková et al., 2010). XRF analysis is an analytical method which is limited to an identification of chemical elements in a volume close to a surface of an object. The thicknesses of analyzed materials depend on a measured element, chemical composition, and experimental conditions. It is usually only several tens of micrometers. The present elements can be distributed heterogeneously even in this thin profile. The most typical examples are effects of corrosion processes, coatings, and other more complicated layered structures.

There are several methods available for identification of a layered structure using gamma and X-ray spectrometry. These include a depth-profiling technique based on at least two measurements performed under different experimental conditions (Trojek and Čechák, 2007), a method based on the detected ratio of at least two lines of an analyte in an investigated object, and a confocal XRF requiring special instrumentation including focusing and collimating X-ray optics (Kanngießer et al., 2004).

This paper deals with the method based on the measurement of a ratio of two gamma or X-ray lines of a radionuclide or a chemical element, respectively. Its principle consists in different attenuation coefficient for these two lines in a measured material. If an analyte is present at some depth, its radiation has to penetrate through thick layer of the matrix, and the photon fluxes are significantly changed. For instance, ^{214}Bi is a uranium progeny and is present in soil or walls of buildings. At least two lines of this radionuclide can be detected with an HPGe detector, and thus net peak area ratio of these lines is related to depth distribution of this radionuclide. Analogously in XRF, ratios of Kα, Kβ, and L lines of an element can provide us with information on depth distribution of this element in an analyzed object. If an element is located deeper in a matter, its Kα/Kβ ratio is usually decreased because the Kα line has lower energy which is generally more absorbed than the line Kβ. This method was successfully applied to identification of pigment layers in illuminated manuscripts (Trojek et al., 2010) and

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paintings (Trojek and Trojková, 2015), thickness measurement of coated Ni on brass of Karimi et al. (2009), and recognition of gold and gilding (Trojek and Hložek, 2012). It was shown that the Kα/Kβ ratio of an element depends also on its concentration (e.g. self-attenuation), and therefore the Kα/Kβ diagrams, displaying Kα/Kβ ratio versus Kα intensity, are preferable than solely Kα/Kβ ratio (Trojek et al., 2008).

This paper shows how the detected net count rate ratios in GS and XRF depend on depth distribution of an analyte, and mainly how these ratios depend on density and chemical composition of measured materials.

2. Gamma spectrometry

2.1. Experimental setup and data analysis

In the case of gamma spectrometry, we expect that presented method, based on different attenuation for different energies, could be useful in environmental monitoring and the investigation of depth distribution of (natural) radionuclides.

All Monte Carlo simulations were implemented using MCNP Code (Pelowitz, 2013) – MCNP6, the latest release of the code. MCNP (Monte Carlo N-Particle) is a general-purpose code which is being developed by Los Alamos National Laboratory. All geometry plots presented in this paper were acquired using VisEd (Visual Editor constructed for MCNP).

The studied geometry consisted of a detector placed 1 m above the ground and the source of radiation, which in this case was a flat ground. The detector model was created according to real scintillation detector (GS256 – NaI(Tl), 3″x 3″). These detectors (or similar ones) are frequently used in environmental in-situ measurements. The visualization of the detector model is in Fig. 1.

The real infinite half-space source (ground) was approximated by half-sphere of 5 m radius. The position of the detector placed above the surface is pictured in Fig. 2. Two different types of radionuclide distribution within the source cell were considered. First type was a homogenous distribution in the whole half-sphere and the second type was a 5 cm thick layer with homogenous distribution in different depths. In the same time, the effect of soil composition and humidity was investigated by running the simulations for standard loam and sand (both in 3 levels, 0%, 10% and 20% of specific humidity), and for granite (bedrock with 0% humidity). The composition and density of those materials were specified according to McConn et al. (2011).

Only radionuclides emitting at least two gamma-ray lines with different energy (with sufficient yield) can be considered in this problem. Since only natural radionuclides are a matter of concern in this paper, four of them were chosen for the simulations and are displayed in Table 1. Each energy line was simulated separately, where the probability of photon emission from the source was set to 100%. In reality, this probability is smaller than 100%, therefore to compare the simulation results with the measurement, the results would need to be multiplied by the yield.

The aim of these simulations was to calculate the probability of detecting a photon emitted within the source by the detection unit.[1] The best choice of tally (=part of the input file where the user specifies what should be calculated) to provide this estimation is F8 tally. Energy window with the center in the simulated energy and width 40 keV was sufficient to calculate photopeak efficiency, since Gaussian energy broadening was not included in the simulations. Using this specification of tally, every simulation gives us the number of impulses in NaI(Tl) crystal in energy window (E-20 keV, E+20 keV), where E is

[1] The detection efficiency was calculated in order to have a possibility for the future to compare the results with the measured value, if some experiment should be carried out in this area. Since only ratios are dealt with in this paper, the calculated value and therefore the tally could be chosen differently with the same result.

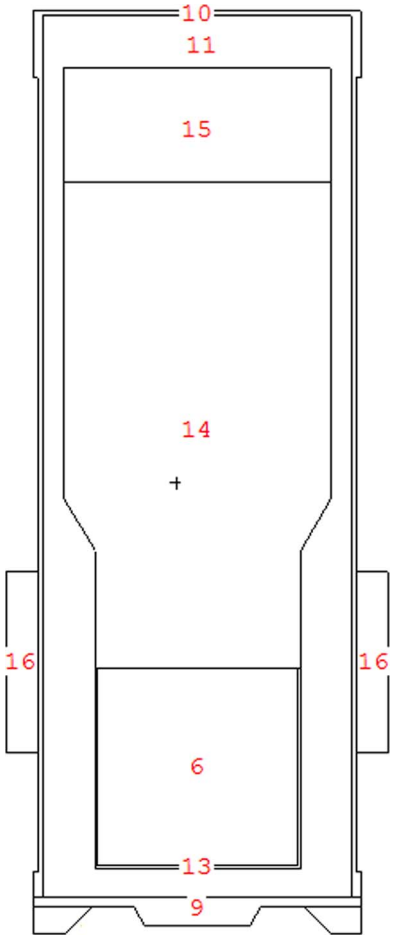

Fig. 1. Model of the scintillation detector. Cell 6 is the NaI(Tl) crystal, cell 14 is photomultiplier and cell 15 is the electronics. The detection unit is protected by Al foil (cell 13) PU foam (cell 11) and dural shielding (cells 9, 10).

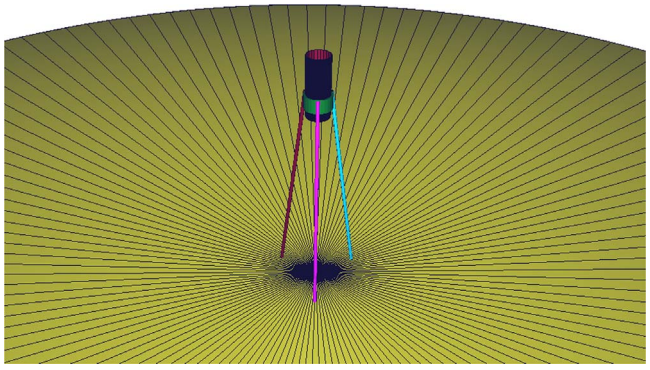

Fig. 2. The detector (on a tripod) placed above the ground.

Table 1
Energy and yields of gamma-ray lines of selected radionuclides suitable for simulation (Gilmore, 2008).

Radionuclide	Energy (keV)	Yield (%)
^{235}U	143.76	10.96
	185.72	57.20
^{214}Pb	295.22	18.50
	351.93	35.60
^{208}Tl	583.19	30.60
	2614.51	35.85
^{214}Bi	609.31	45.49
	1764.49	15.28