



Application of neural network method to detect type of uranium contamination by estimation of activity ratio in environmental alpha spectra



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ABSTRACT

The discrimination of the composition of environmental and non-environmental materials by the estimation of the $^{234}\text{U}/^{238}\text{U}$ activity ratio in alpha-particle spectrometry is important in many applications. If the interfering elements are not completely separated from the uranium, they can interfere with the determination of ^{234}U . Thickness as a result of the existence of iron in the source preparation phase and their alpha lines can broaden the alpha line of ^{234}U in alpha spectra. Therefore, the asymmetric broadening of the alpha line of ^{234}U and overlapping of peaks make the analysis of the alpha particle spectra and the interpretation of the results difficult. Applying Artificial Neural Network (ANN) to a spectrometry system is a good idea because it eliminates limitations of classical approaches by extracting the desired information from the input data. In this work, the average of a partial uranium raw spectrum, were considered. Each point that its slope was of the order of 0–1% per 10 channels, was used as input to the multi-layer feed forward error-back propagation network. The network was trained by an alpha spectrum library which has been developed in the present work. The training data in this study was actual spectral data with any reasonable thickness and interfering elements. According to the results, the method applied to estimate the activity ratio in this work, can examine the alpha spectrum for peaks which would not be expected for a source of given element and provide the clues about composition of uranium contamination in the environmental samples in a fast screening and classifying procedures.

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1. Introduction

The measurement of the activity of radionuclides and their ratios are very interesting for determining the composition of environmental and non-environmental materials in many applications including nuclear power industry, radioactive waste management and disposal, health physics, geology, geochronology, and environmental science (Fleischer, 2008; Minteer et al., 2007; Rubio Montero et al., 2004). Among analytical methods for isotopic determination of radionuclides, Alpha-particle spectrometry with silicon detectors is known as an appropriate, precise, and low cost technique to assay alpha-particle emitting elements such as Uranium isotopes (Jia et al., 2005; García-Torano, 2006; Kunzendorf, 1968; Minteer et al., 2007). The activity ratios, for example in

nuclear power industry, depend on the extent of the enrichment process, which have caused depletion in the lighter uranium isotopes and can be used for distinguishing natural from anthropogenic sources of uranium (Minteer et al., 2007). For natural uranium, the $^{234}\text{U}/^{238}\text{U}$ activity ratio in soil typically ranges from 0.5 to 1.2 (Sansone et al., 2001; Minteer et al., 2007). The corresponding $^{235}\text{U}/^{238}\text{U}$ activity ratio is approximately 0.046. Depleted uranium has lower $^{234}\text{U}/^{238}\text{U}$ and $^{235}\text{U}/^{238}\text{U}$ activity ratios. For an isotopic abundance of 0.2% for ^{235}U , these ratios become 0.18 ($^{234}\text{U}/^{238}\text{U}$) and 0.013 ($^{235}\text{U}/^{238}\text{U}$) respectively (Sansone et al., 2001). Conversely, $^{234}\text{U}/^{238}\text{U}$ and $^{235}\text{U}/^{238}\text{U}$ activity ratios are more than 1.2 and 0.046, respectively, in enriched uranium (Minteer et al., 2007). Experimental determination of the activity ratios between ^{234}U , ^{235}U and ^{238}U are affected by uncertainties, the magnitude of which depend upon the accuracy and precision of the measurements of the single isotopes. For example, a counting time of 168 h was necessary to measure the ^{235}U concentration and its activity ratio ($^{235}\text{U}/^{238}\text{U}$) in alpha spectrometry, due to its lower activity concentration and associate with the high counting

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uncertainty (Jia et al., 2005; Desideri et al., 2002). But, the natural abundance of ^{238}U and specific activity of ^{234}U , demonstrate that their activity concentration and activity ratio directly can be measurable by alpha spectrometry. As an inference from these facts, the alpha spectrometry is a technique that allows the independent and more accurate measurement of the two uranium isotopes and their ratio (Minteer et al., 2007). Therefore, the values of the activity ratio $^{234}\text{U}/^{238}\text{U}$, as a qualitative indicator for the presence natural, depleted or enriched uranium, permit discrimination between natural and anthropogenic source of uranium (Minteer et al., 2007; Jia et al., 2005). However, the interfering elements, such as: Iron (Fe), radium (^{226}Ra), thorium (^{230}Th) and protactinium (^{231}Pa), are not completely separated from the uranium and can interfere with the determination of ^{234}U (ASTM C1000-11). Thickness as a result of the existence of iron in the source preparation phase and their alpha lines can broaden the alpha line of ^{234}U in alpha spectra. Therefore, the asymmetric broadening of the alpha line of ^{234}U and overlapping of peaks make the analysis of the alpha particle spectra and the interpretation of the results difficult (Minteer et al., 2007; Sánchez and Montero, 1999).

To resolve this problem, de-convolution processes are often used based on the semi-empirical functions of shape of a mono-energetic alpha peak with a large number of fitting parameters (Sánchez and Montero, 1999). Their physical meaning is obscure or completely lacking. In particular, for environmental measurements which are usually low-level counting, this excessive number of fitting parameters can give rise to anomalies in the values obtained for some of them (Baeza et al., 2011). Also, the use of collimator can decrease the low energy tail and the energy distribution broadening, but the detection efficiency is sacrificed (Park et al., 2014; Pöllänen et al., 2013; Pöllänen et al., 2012).

An alternative method to resolve these problems is the usage of Artificial Neural Network (ANN) method. The main advantage of ANN is that, it does not need any mathematical model since an ANN learns from examples and recognizes patterns in a series of input and output data without any prior assumptions about their nature and interrelations (Basheer and Hajmeer, 2000). ANN eliminates the limitations of classical approaches by extracting the desired information from the input data. Applying ANN to a spectrometry system needs sufficient input and output data instead of mathematical equations for performing the fit to nuclear spectra, including X-, gamma-ray and alpha-particles spectra (Baeza et al., 2011; Basheer and Hajmeer, 2000; Keller et al., 1995; Yoshida et al., 2002; Kangas et al., 2008; Chen and Wei, 2009; Medhat, 2012; Miranda et al., 2009; Doostmohammadi et al., 2010). For each nuclear spectrum, such as alpha spectrum, up to 2048 data points are selected as inputs. The numerous inputs increase the complexity of a network, which is not desirable. It leads to slow down the convergence in training and degrade the generalization.

The scope of the paper is to improve of the performance of alpha-particles spectrometry in the spectrum analysis in order to identify the type of contamination in the soil samples containing uranium. The Multilayer feed forward error-back propagation neural network, as an estimating tool, is used to estimate the $^{234}\text{U}/^{238}\text{U}$ activity ratio, because it is a good choice to screen and distinguish between natural and anthropogenic spectra in a number of applications (Keller et al., 1995; Yoshida et al., 2002; Minteer et al., 2007; Kangas et al., 2008; Doostmohammadi et al., 2010; Medhat, 2012). The network was trained by an alpha spectrum library which has been developed in the present work. To reduce the input data size, instead of using a feature extraction technique such as Principle Component Analysis (PCA) (Li et al., 2008), a partial uranium spectrum from 3.5 MeV to 5.5 MeV of the raw spectrum was selected and an average of 2 consecutive channels, as the new

value of the first channel, was calculated and plotted as the average curve. Each point that its slope was of the order of 0–1% per 10 channels, was used as input to the networks. After training and demonstrating the capability of the networks to estimate the activity ratio, the verification of the network was evaluated according to statistical indexes including: the Coefficient of Correlation (CC), Mean Absolute Prediction Error (MAPE) and the Root Mean Square Error (RMSE).

2. Materials and methods

2.1. Source preparation

The radiochemical method for determination of uranium isotopes in different compositions of soil samples is ASTM C1000-11 method (ASTM C1000-11). In this procedure, a soil sample (10 g) was mixed with U-232 tracer (SMR 4324A, NIST, USA). It was heated to destroy organic materials and dissolved with a mixture of hydrofluoric acid and nitric acid (Merck, Germany). The uranium was co-precipitated with ferric hydroxide (Merck, Germany) and the precipitate was dissolved with hydrochloric acid (Merck, Germany). Iron was removed by the extraction with isopropyl ether (Merck, Germany). Radium and thorium were separated from uranium by anion exchange (Dowex 1×8 , Cl-form, 100–200 mesh, Sigma chemical company, USA). Uranium was electrodeposited on a stainless steel disk and determined by alpha spectrometry. In this work, 125 uranium sources of environmental soil samples were prepared for the real library with any reasonable thickness and interfering elements. It was 40 percent depleted uranium and 60 percent natural uranium.

Alpha-particle spectra were obtained by measuring the sources with a passivated ion – implanted planar silicon detector with a 450 cm^2 active area (14 keV nominal resolution) mounted in a vacuum chamber and coupled to a low-noise preamplifier, a pulse-shaping amplifier, and a PC-resident multichannel analyzer (2048 channels). The source-to-detector distance was 7 mm ($\epsilon \approx 22\%$). The counting time was 1–3 days. The uncertainty was 1σ for individual analysis (the activity of the uranium isotopes (^{234}U and ^{238}U) and their activity ratio values in the samples). It was estimated from the uncertainties associated with the tracer activity, the addition of tracer to the sample and the counting statistics of the sample and the blank, etc. The ^{238}U activity concentration and activity ratio, chemical yield ranges in the samples were $3.5\text{--}55 \text{ Bq.kg}^{-1}$, $0.10\text{--}1.2$, $48\text{--}67\%$ respectively.

It should be mentioned that, the activity of some samples that were separated completely from the interfering elements and were not thick, was determined by the sum of area under peak, directly. But, the samples that were not separated completely from the interfering elements or were thick, their peak area was estimated by using the fitting techniques (Sánchez and Montero, 1999) and the activity was calculated.

2.2. Reduction of the input data size

Artificial neural network can handle small input data sets better. To reduce the amount of input data, Keller et al. (Keller et al., 1995) adopted an average of 10 consecutive channels to feed into the ANN. They have reduced the original 512 channels to 20 channels for the analysis of alpha spectra. Pilato et al. (Pilato et al., 1999) used 12 zones of interesting gamma-ray peak regions in a 2k-channel spectrum. Medhat and Yoshida et al. (Yoshida et al., 2002; Medhat, 2012) used only peak channels corresponding to gamma-ray peak energy as the input data. In this work, firstly, a partial uranium spectrum from 3.5 MeV to 5.5 MeV of the raw spectrum was selected. A typical uranium spectrum, for example, is shown in

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