Applied Radiation and Isotopes 105 (2015) 225–232

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Evaluation of the suitability of neural network method for prediction of uranium activity ratio in environmental alpha spectra

Mohammad Reza Einian, Seyed Mahmood Reza Aghamiri *, Reza Ghaderi

Department of Radiation Medicine, Shahid Beheshti University, P.O. Box 1983963113, Tehran, Iran

HIGHLIGHTS

- The suitability of RBF network method for the prediction of $^{234}U/^{238}U$ activity ratio was evaluated.
- The average of 2 consecutive channels of a partial uranium raw spectrum was plotted as the average curve.
- The points that their slopes are of the order of 0–1% per 10 channels, were used as inputs to the RBF.
- The network was trained by the simulated spectra library, could accurately predict the activity ratio.
- The network was trained by the real spectra library, could reasonably predict the activity ratio.

ARTICLE INFO

Article history:
Received 29 November 2014
Received in revised form
15 July 2015
Accepted 14 August 2015
Available online 15 August 2015

Keywords:
Alpha spectrometry
Activity ratio
Radial Basis Function neural network
Advanced Alpha Spectrometric Simulation (AASI)
Depleted uranium

ABSTRACT

Applying Artificial Neural Network to an alpha spectrometry system is a good idea to discriminate the composition of environmental and non-environmental materials by the estimation of the $^{234}U/^{238}U$ activity ratio. Because it eliminates limitations of classical approaches by the extraction the desired information from the average of a partial uranium raw spectrum. The network was trained by an alpha spectrum library which was developed in this work. The results indicated that there was a small difference between the target values and the predictions. These results were acceptable, because the thickness of samples and the inferring elements were different in the real library.

1. Introduction

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-Toraño, 2006; Kunzendorf, 1968). The measurement of the alpha-activity ratios of uranium isotopes is very interesting for determining the composition of environmental and non-environmental materials in many applications including nuclear industry, radioactive waste management and disposal, health physics, geology, geochronology, and environmental science (Rubio Montero et al., 2004). The activity ratio depends on the extent of the enrichment process, which has caused depletion in the lighter uranium isotopes and can be used for distinguishing natural from anthropogenic sources of uranium. For natural uranium, the $^{234}U/^{238}U$ activity ratio in soil typically ranges from 0.5 to 1.2 (Sansone et al., 2001). The corresponding $^{235}U/^{238}U$ activity ratio is approximately 0.046. Depleted uranium has lower $^{234}U/^{238}U$ and $^{235}U/^{238}U$ activity ratios. For an isotopic abundance of 0.2% for ^{235}U, these ratios become 0.18 ($^{234}U/^{238}U$) and 0.013 ($^{235}U/^{238}U$) respectively (Sansone et al., 2001). 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 in alpha spectrometry to measure the ^{235}U concentration and the activity ratio $^{235}U/^{238}U$, due to its lower activity concentration and associate with the high counting 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

* Corresponding author.
E-mail addresses: M_einian@sbu.ac.ir (M.R. Einian), smr-aghamiri@sbu.ac.ir (S.M.R. Aghamiri), R_ghaderi@sbu.ac.ir (R. Ghaderi).

http://dx.doi.org/10.1016/j.apradiso.2015.08.017
0969-8043/

spectrometry is a technique that allows the independent and more accurate measurement of the two uranium isotopes and their ratio. Therefore, the values of the activity ratio $^{234}U/^{238}U$ permit discrimination between natural and anthropogenic source of uranium (Jia et al., 2005). However, there are difficulties to analyze the environmental samples with a complex matrix composition. The main reason for difficulties is the existence of the interfering elements such as: radium (^{226}Ra), thorium (^{230}Th) and protactinium (^{231}Pa) in a complex matrix composition. They are not completely separated from the uranium and can interfere with the determination of ^{234}U (ASTM C1000-11). 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 difficult (Sánchez and Montero, 1999).

To resolve this problem, de-convolution processes are often used based on the semi-empirical functions of the 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).

An alternative method to resolve these problems is the use 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.

In this work, in order to evaluate the suitability of neural network method for prediction of uranium activity ratio, the Radial Basis Function (RBF) Neural network was applied to estimate the $^{234}U/^{238}U$ activity ratios, because it is a good choice to screen and distinguish between natural and anthropogenic uranium in a number of alpha spectra. 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. The points that slopes are of the order of 0–1% per 10 channels were used as inputs to the RBF network. The RBF network was trained by an alpha spectrum library which has been developed in the present work. Since the obtaining of library spectra in a laboratory is usually expensive and time consuming, at first, a Monte Carlo method was utilized. The library produced by a set of thin and uniform uranium sources, was used as the input of the RBF network. By using of Monte Carlo method known as Advanced Alpha Spectrometric Simulation (AASI) (Siiskonen and Pöllänen, 2005) and electrodeposited source characteristics, a large number of alpha-particles spectra were produced. The AASI 2.0 computer code has been developed to simulate alpha particle energy in alpha spectrometry (Siiskonen and Pöllänen, 2005). The AASI has been validated against measurements, analytical calculations, and other simulation codes (Siiskonen and Pöllänen, 2004; Pöllänen et al., 2005; Pöllänen and Siiskonen, 2006; Pöllänen et al., 2007; Ranebo et al., 2010). After demonstrating the capability of the network to predict the activity ratio, the real library was produced by a set of the uranium sources. They have to be prepared by electro deposition which is the technique used for the source preparation (Talvitie, 1972; Hallstadius, 1984; Beesley et al., 2009; Méndez et al., 2010).

1.1. Principle of the method

Artificial neural networks can best handle small input data sets. To reduce the amount of input data, 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. (1999) used 12 zones of interesting gamma-ray peak regions in a 2k-channel spectrum. Medhat (2012) and Yoshida et al. (2002) 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 Fig. 1a. In this figure, three peaks showing the existence of uranium isotopes ^{238}U, ^{234}U, and ^{232}U are clearly visible. An average of 2 consecutive channels, as the new value of the first channel, is calculated using the following equation:

$$A_i = 0.5 \times (\mathrm{Count}_{i+1} + \mathrm{Count}_i) \quad (1)$$

where i is the number of channel, A_i is the average of 2 consecutive channels, Count_{i+1} and Count_i are the counts in $(i+1)$-th channel and i-th channel, respectively.

After the calculation, a collection of points are produced that arranged in the manner of a series of steps (Fig. 1b). The new curve is similar to the integral pulse high distribution. The abscissa in the new curve is the same abscissa of the spectrum. The ordinate now represents the average of 2 consecutive channels. Where peaks appear in the spectrum, such as the alpha peaks of U-238 and U-234, the increase will occur in the magnitude of the slope of the average curve. On the other hand, where peaks do not appear (such as A, B, C and D points in Fig. 1b) regions of minimum magnitude of the slope are observed in the average curve. It is desirable to select the points, as input data-set, that will provide the minimum magnitude of the slope in the curve.

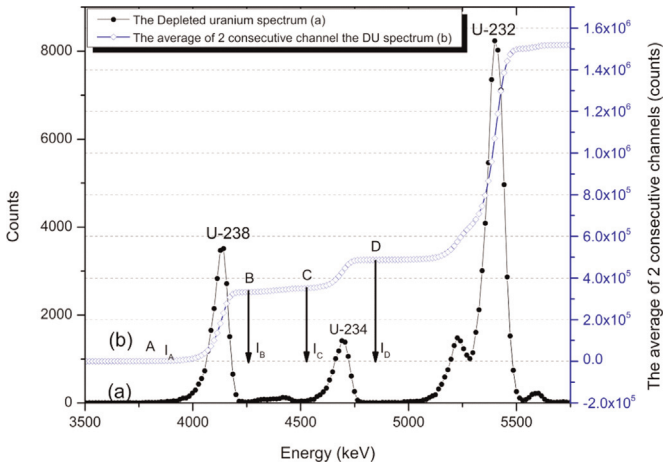

Fig. 1. An example of the spectrum from a depleted uranium sample and its tracer: (a) original spectrum, (b) The average of 2 consecutive channels. IA, IB, IC and ID are the ordinate values of points A, B, C and D, respectively, that their slopes were of the order of 0–1% per 10 channels from regions of 3.5 to 4.0 MeV, 4.2 to 4.3 MeV, 4.4 to 4.6 MeV and 4.8 to 5.0 MeV in the average curve.