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A new approach to the analysis of alpha spectra based on neural network techniques

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ABSTRACT

The analysis of alpha spectra requires good radiochemical procedures in order to obtain well differentiated alpha peaks in the spectrum, and the easiest way to analyze them is by directly summing the counts obtained in the Regions of Interest (ROIs). However, the low-energy tails of the alpha peaks frequently make this simple approach unworkable because some peaks partially overlap. Many fitting procedures have been proposed to solve this problem, most of them based on semiempirical mathematical functions that emulate the shape of a theoretical alpha peak. The main drawback of these methods is that the great number of fitting parameters used means that their physical meaning is obscure or completely lacking. We propose another approach—the application of an artificial neural network. Instead of fitting the experimental data to a mathematical function, the fit is carried out by an artificial neural network (ANN) that has previously been trained to model the shape of an alpha peak using as training patterns several polonium spectra obtained from actual samples analyzed in our laboratory. In this sense, the ANN is able to learn the shape of an actual alpha peak. We have designed such an ANN as a feed-forward multi-layer perceptron with supervised training based on a back-propagation algorithm. The fitting procedure is based on the experimental observables that are characteristic of alpha peaks-the number of counts of the maximum and several peak widths at different heights. Polonium isotope spectra were selected because the alpha peaks corresponding to ²⁰⁸Po, ²⁰⁹Po, and ²¹⁰Po are monoenergetic and well separated. The uncertainties introduced by this fitting procedure were less than the counting uncertainties. This new approach was applied to the problem of resolving overlapping peaks. Firstly, a theoretical study was carried out by artificially overlapping alpha peaks from actual samples in order to test the ability of the ANN to resolve each peak. Then, the ANN procedure was checked by determining the activity levels of different spectra obtained from certified samples for which one knows a priori the radioactive content, and its results were compared with those of other methods.

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1. Introduction

The determination of the alpha emitting radionuclide content of environmental samples requires a prior radiochemical procedure so as to produce an alpha source as thin and free of interferences as possible. The principal reason is to reduce the degradation of the shape of the alpha peaks in the low-energy tails due to selfabsorption in the sample. The use of a vacuum between source and detector also reduces the problem of energy losses of the alpha particles. Despite such precautions, however, low-energy tails are frequently observed in the alpha spectra obtained from environmental samples. If the alpha peaks obtained in the spectra are well

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separated in energy, the easiest way to determine the activity level is by directly summing the counts that occurred in all the channels that define the peak. But, in environmental samples, the lowenergy tails of the higher energy alpha peaks usually overlap partially with other peaks of lower energy. Many methods have been proposed to solve the overlap problem, most of them based on semi-empirical mathematical functions that emulate the shape of a theoretical alpha peak. Examples are the proposals of Bortels and Collaers [1] and Westmeier and Van Aarle [2], whose functional forms were capable of describing very well the shape of the peaks obtained with silicon semiconductor detectors. The main drawback of such methods is that the great number of fitting parameters used means that their physical meaning is obscure or completely lacking. For environmental measurements in particular, which are usually low-level counting, this excessive number of fitting parameters can also give rise to anomalies in the values that

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Fig. 1. Scheme of the artificial neural network used with three layers of neurons: input, hidden, and output.

are obtained for some of them [3]. To resolve these problems, we took a radically different approach—applying an artificial neural network (ANN) that had previously been trained to reproduce the shape of an alpha peak, i.e., the ANN learns the shape of a real alpha peak from real spectra obtained in the laboratory. An ANN is an artificial intelligence tool that identifies arbitrary nonlinear multiparametric discriminant functions directly from experimental data. It simulates the structure and/or functional aspects of biological neural networks, and they are frequently used to model complex relationships between inputs and outputs or to find patterns in data. Fig. 1 shows a general schema of an ANN with three layers of neurons: input, hidden, and output layer.

2. Design of the artificial neural network (ANN)

We designed the ANN as a feed-forward multi-layer perceptron with supervised training based on a back-propagation algorithm. The fitting procedure is based on the experimental observables that are characteristic of alpha peaks—the number of counts of the maximum and different peak widths at several heights (see Fig. 2), which were taken as the ANN's input parameters. The spectra supplied to the ANN were selected from polonium samples analyzed in our laboratory as part of routine and other research work. The reason for this selection was because the alpha peaks corresponding to ²⁰⁸Po, ²⁰⁹Po, and ²¹⁰Po are monoenergetic and well separated. These spectra had also been acquired by different detectors. The generalization of the shape of an alpha peak obtained by the ANN was therefore independent of the radionuclide considered and of the detector in which the spectrum was acquired. The network was trained on more than 300 different polonium alpha peaks, using a crossvalidation method with a mean squared error (MSE) cost function to determine the training level. The network training method has



Fig. 2. Alpha peak characteristics selected as the input parameters to the ANN. 1: maximum height; 2: right half of width at 4/5 maximum; 3: left half of width at 4/5 maximum; 4: right half of width at 2/3 maximum; 5: left half of width at 2/3 maximum; 6: right half of FWHM; 7: left half of FWHM; 8: right half of width at 1/3 maximum; 9: ROI width.

been explained elsewhere [4]. The output of the ANN is the number of counts for each channel corresponding to the ROI of the initial alpha peak according to the shape the ANN learned during its training.

3. ANN fitting procedure

The uncertainties introduced by this fitting procedure were typically less than the counting uncertainties associated with the determination of the integral of the alpha peak [4]. Fig. 3 shows by way of example three ANN fits of polonium alpha peaks. Table 1 presents the comparisons of the conventional direct sums of these spectra with the integrals of the ANN fits, and of the ANN fits with a functional shape of the alpha peak (ALFIT in the table) [2]. One observes that the ANN fit was more accurate for the low-level counting.

This new approach was applied to the problem of resolving partially overlapping peaks in two steps. Firstly, a theoretical study was carried out by artificially overlapping alpha peaks from actual samples in order to test the ability of the ANN to resolve each peak, and to uncover any limitations that the procedure might have [5]. The first step of the procedure to solve partially overlapped peaks of a spectrum was to extract the characteristics of the higher energy peak, located on the right side in the spectrum. This information was then fed to the neural network. We used the neural network output to fit that peak and subtract it from the original spectrum. The result was the isolated left peak, which could then be analyzed using the single peak fitting procedure again. The ANN predictions for each overlapped peak usually were within the counting uncertainties. The ANN fit was also checked by determining the activity levels of different spectra obtained from certified samples in which one knows a priori the radioactive content, and its results were compared with those of other more conventional methods. The results of this comparison for various radionuclides and reference materials with different matrices are presented in Table 2. There was good agreement with the activities calculated by means of direct sums, and more importantly, the values lie within the 95% confidence intervals corresponding to those radionuclides.

Finally, the ANN procedure was applied to an actual uranium spectrum obtained in our laboratory from a surface water sample (Fig. 4). Different alpha peaks corresponding to a given radionuclide were considered—²³⁸U: 4196 and 4147 keV; ²³⁴U:



Fig. 3. Result of the fit of an alpha peak using the ANN. Solid bars: Experimental alpha peak obtained in the laboratory. Solid line: Output alpha peak shape from the ANN.

Table 1

Values of the integrals corresponding to alpha peaks A, B, and C in Fig. 2 calculated as direct sums, their corresponding counting uncertainties, and the difference between the direct sum and the ANN fit and ALFIT [2,3], expressed as percentages.

Peak	Direct sum	Counting uncertainty (%)	Difference with ANN fitting (%)	Difference with ALFIT (%)
А	124 ± 11	8.98	0.80	4.03
В	304 ± 17	5.73	0.33	25
С	532 ± 23	4.34	3.38	0.19

4775.8 and 4723.7 keV; and 232 U: 5320.3 and 5263.5 keV. The integrals of those peaks calculated as direct sums were 130, 141, and 1197 counts for 238 U, 234 U, and 232 U, respectively, whereas

the ANN fit gave 95, 132, and 1252 counts, respectively. As it can also be seen in Fig. 4 there was a good agreement between the actual spectrum and the ANN fitting, with never more than a 15-count difference between the number of counts observed in a channel of the spectrum and the ANN fitting.

4. Conclusions

The use of an artificial neural network to fit alpha peaks allows the frequently low-level alpha spectra obtained from environmental samples to be analyzed with precisions within the counting uncertainties for a single alpha peak. It was also able to resolve the overlapping alpha peaks corresponding to the same radionuclides, such as ^{232,234,238}U. The ANN fitting procedure used

Table 2

Activity level of ^{238,234}U and ²¹⁰Po in reference materials, calculated as direct sums and from the ANN fit.

Reference material	Radionuclide	Recommended value (range) (Bq/kg)	Activity from direct sum (Bq/kg)	Activity from ANN fitting (Bq/kg)
IAEA-381 (Sea water)	²³⁴ U	0.050 (0.0443-0.058)	0.048 ± 0.009	0.052 ± 0.009
	²³⁸ U	0.041 (0.038-0.048)	0.041 ± 0.009	0.047 ± 0.009
IAEA-327 (Soil)	²¹⁰ Po	58.8 (53.9-63.7)	61 ± 16	61 ± 16



Fig. 4. Uranium spectrum obtained from a surface water sample. The goodness of the fit is shown below as the difference between the counts of the actual spectrum and the alpha fitting prediction.

directly measurable characteristics of the alpha peaks in a spectrum as input parameters, i.e., there is no "artificial" fitting parameter with a potentially dubious physical meaning. the pre-doctoral formation for researchers (D.O.E. 130/2007)", and for financial support to the LARUEX research group (FQM001).

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