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Application of neural networks for the analysis of gamma-ray spectra measured with a Ge spectrometer

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Abstract

The analysis of gamma-ray spectra to identify lines and their intensities usually requires expert knowledge and time-consuming calculations with complex fitting functions. A neural network algorithm can be applied to a gamma-ray spectral analysis owing to its excellent pattern recognition characteristics. However, a gamma-ray spectrum typically having 4096 channels is too large as a typical input data size for a neural network. We show that by applying a suitable peak search procedure, gamma-ray data can be reduced to peak energy data, which can be easily managed as input by neural networks. The method was applied to the analysis of gamma-ray spectra composed of mixed radioisotopes and the spectra of uranium ores. Radioisotope identification was successfully achieved. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Gamma-ray spectrometry; Neural network; Radioisotope identification

1. Introduction

An artificial neural network (ANN) algorithm is now being used in a wide variety of data processing applications such as predictions, non-linear problems and real time data analysis. An advantage of this technique is rapid pattern recognition with an appropriate training in advance. This technique can also be applied in the field of nuclear spectroscopy, e.g. resolution improvement of CdZnTe detectors employing pulse-shape treatment [1] and X-, gamma- and

alpha-ray spectrometry [2–7]. However, it is rarely used in gamma-ray spectrometry obtained by Ge detectors [8,9]. This is because the spectral size, which is typically 4096 channels or more, is too large as an input data size for the ANN. Generally, a gamma-ray spectrum is composed of a number of photo peaks from several radioisotopes and the associated Compton continuum. Spectral analysis needs functional fitting of complex peaks, a time-consuming procedure requiring expert knowledge. One purpose of spectral analysis is to identify the radionuclide included in the sample and to determine the amount of radioactivity. Once radionuclide identification has been accomplished, the activity can be deduced from the peak counting rate by correcting for the detection efficiency.

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In this work, the ANN was applied to qualitative analysis of gamma-ray spectra. To reduce the input data size, only peak energy information was processed as input data, and patterns of emitted gamma-ray energies from individual nuclides were used for the initial training procedure. An ANN model with a back-propagation (the following BP) algorithm was used for the training. The present ANN analysis was applied to the gamma-ray spectra of standard sources and uranium ore [10].

2. Method of neural network approach

2.1. Neural networks

Neural network computing is analogous to biological neural systems. The ANN is composed of an input layer, one or more hidden layers (no hidden layer is also possible) and an output layer. Each layer includes neurons which are connected to all the neurons of a successive layer. Each neuron has its own weight and the sum of the weights is processed with a transformation such as a sigmoid function.

The weights of each neuron are established through a training procedure. The training of the ANN can be performed with a BP algorithm. In this algorithm, a number of example data whose outputs are known are used as input data. The calculated output is then compared with the known output. The difference between the two outputs is then backpropagated to recalculate the weights. Such an iterative procedure is continued until the difference becomes desirably small.

2.2. Principle of the present method

ANNs can best handle small input data sets. To reduce the amount of input data, Kangas et al. [5] 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. [8] used 12 zones of interesting gamma-ray peak regions in a 2K-channel spectrum. The principle of the present work is to use only peak channels corresponding

to gamma-ray peak energy as the input data. The input data size becomes at most one hundred data points, which can be handled by the ANN.

First, a supervised data set composed of 53 gamma-ray energies was selected from 28 radioisotopes. These radioisotopes were typical gamma-ray standard sources and natural radioisotopes such as uranium, actinium and thorium series. Only dominant gamma-rays from these radioisotopes were adopted for the supervised data set. Radioisotopes and gamma-ray energies adopted for the supervised data set are given in Table 1.

Table 1
Supervised data set composed of 28 radioisotopes and 53 gamma-ray energies

| Radioisotope | Energy (keV) |
|------------------------|--|
| ²² Na | 1274.54 |
| ⁴⁰ K | 1460.8 |
| ⁴⁶ Sc | 889.25, 1120.51 |
| ⁵¹ Cr | 320.08 |
| ⁵⁴ Mn | 834.83 |
| ⁵⁶ Co | 846.75, 1238.26 |
| ⁵⁷ Co | 122.06, 136.47 |
| ⁵⁸ Co | 810.76 |
| ⁶⁰ Co | 1173.24, 1332.50 |
| ⁸⁸ Y | 898.02, 1836.0 |
| ¹⁰⁹ Cd | 88.03 |
| ¹¹¹ In | 171.28, 245.35 |
| ¹³³ Ba | 81.00, 276.40, 302.85, 356.0, 383.85 |
| ¹³⁷ Cs | 661.65 |
| ¹³⁹ Ce | 165.85 |
| ¹⁵² Eu | 121.78, 344.28, 1408.01 |
| ¹⁵⁴ Eu | 123.14, 723.30, 1274.45 |
| ²⁰⁷ Bi | 569.67, 1063.62 |
| ²⁴¹ Am | 59.54 |
| <i>Uranium series</i> | |
| ^{234m} Pa | 1001.03 |
| ²¹⁴ Pb | 295.21, 351.92 |
| ²¹⁴ Bi | 609.3, 1120.29, 1238.1, 1407.98, 1764.49 |
| <i>Thorium series</i> | |
| ²²⁸ Ac | 338.4, 911.1, 968.9 |
| ²¹² Bi | 727.17 |
| ²⁰⁸ Tl | 583.14, 860.37, 2614.5 |
| <i>Actinium series</i> | |
| ²³⁵ U | 143.76, 185.72 |
| ²²³ Ra | 269.4 |
| ²²⁷ Th | 236.0 |

These energies were provided to each neuron of the input layer of the ANN.

Second, a peak search was carried out of the original gamma-ray spectrum to prepare an input data set. To identify the peak position, the spectrum was smoothed and differentiated twice. Let $n(i)$ is the number of counts in channel number i and its statistical error is $\sigma(i) = \sqrt{n(i)}$. When the second derivative $n''(i)$ satisfied the following equation, a peak position was identified:

$$|n''(i)| > S\sigma''(i)$$

where S is a constant number representing the sensitivity (typically, $S = 3$ was adopted). Following identification, the peak position was converted to the corresponding gamma-ray energy. When the peak energy agrees with one of the supervised data within ± 1.5 keV, the value of this neuron in the input layer is set to 1, otherwise the value is zero. Then, the input data were processed to the ANN. The structure of the present ANN was composed of 47 input layers, 52 hidden layers and 28 output layers as shown in Fig. 1. The number of neurons in the hidden layer was determined through trial and error to be optimal. Each output neuron corresponds to one radioisotope and the output value represents a probability of existence of the radioisotope.

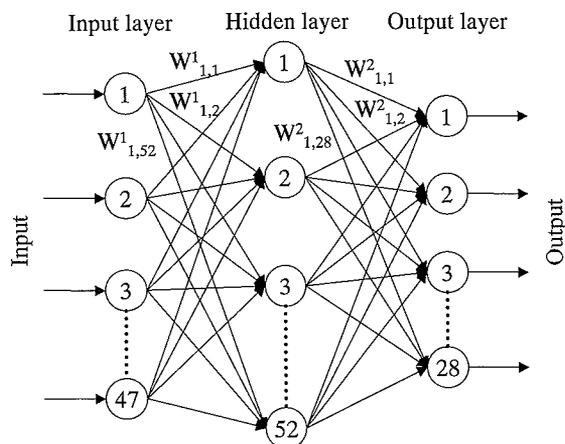


Fig. 1. Structure of ANN used in gamma-ray spectrum analysis.

2.3. Training of the ANN

The training of the present ANN was performed using a number of input data sets which contained an arbitrary number of radioisotopes in the supervised data. Such an input data set is, for example, where the neurons of certain radioisotopes are 1 and the other inputs are zero. The ANN was trained for a total of 409 input data sets. Training input data sets are (i) single radioisotopes comprising 28 sets, (ii) two radioisotopes comprising 378 sets, (iii) three radioisotopes comprising 3 sets. (uranium, thorium and actinium series, respectively). The total iterative frequency was 50,000 and the training time was about 2 h on an Intel 466 MHz celeron processor.

3. Application to gamma-ray spectrum analysis

3.1. Spectral analysis of standard sources

The ANN algorithm was applied to the analysis of three spectra including the gamma-rays of four radioisotopes. There were three combinations of source content as follows: Set 1: ^{54}Mn , ^{60}Co , ^{88}Y , ^{137}Cs ; Set 2: ^{22}Na , ^{88}Y , ^{133}Ba , ^{137}Cs ; Set 3: ^{22}Na , ^{57}Co , ^{88}Y , ^{137}Cs . These sources are gamma-reference point sources (Amersham Int., QCR11) with each activity being 3.7×10^4 Bq. Four sources were arranged at 5 cm from the detector end cap and measurements were continued for 1 h. The HPGe detector (EG&G ORTEC GMX-30200-P, crystal: 63 mm (length) and 58 mm (diameter), resolution: 1.85 keV at 1.33 MeV) was shielded with 20-cm-thick lead bricks. Three gamma-ray spectra are shown in Fig. 2.

An example of the peak search procedure for the sample set 2 is demonstrated in Fig. 3. A partial spectrum from 300 to 500 channels of the raw spectrum, is shown in Fig. 3(a), and the first and second derivatives are shown in Fig. 3(b) and (c), respectively. The output of the ANN is given in Table 2 and depicted as a histogram in Fig. 4. Four radioisotopes in each spectra were exactly identified since the output of the ANN exceeded 0.8 for these radioisotopes. Therefore, the identification of radioisotopes was successfully performed for these spectra.

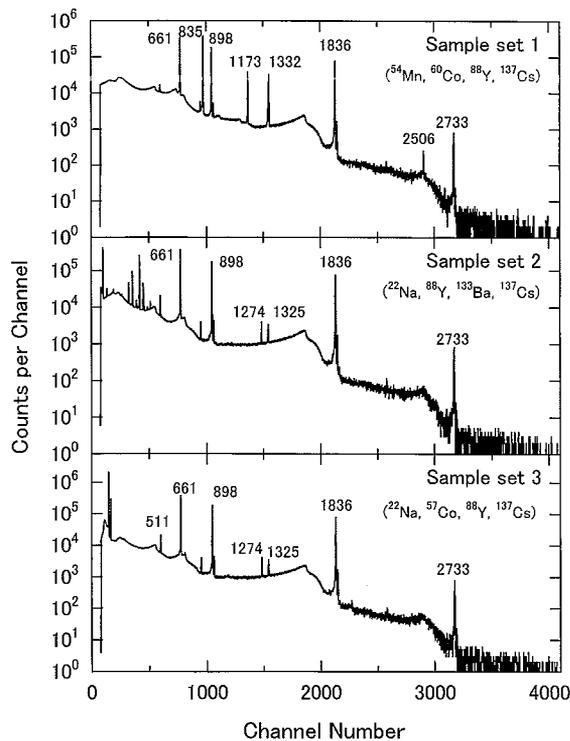


Fig. 2. Gamma-ray spectra obtained from three sets of mixed radioisotopes.

3.2. Radioisotope identification in uranium ore

The identification of gamma-ray spectra of uranium ores is much more complicated because the parent nuclei ^{238}U and ^{235}U have quite long half-lives and their progeny nuclei are in radioactive equilibrium with the parents. As a result, a number of gamma-ray peaks are included in the spectrum. Vigneron et al. [11] applied an ANN to determine the uranium enrichment ratios of UO_2 standard pellets employing a non-destructive method. They analysed KX-rays in the energy range 90–98 keV.

The present ANN algorithm was applied to the analysis of gamma-ray spectra of two uranium ores to identify the radionuclides included in them. These uranium ores are sampled from the natural reactor zone at the Oklo mine. A natural reactor is a phenomenon wherein nuclear fission occurred about 2×10^9 years ago in natural uranium ore without artificial influence. Results of mass-spectrometry are reported by Hidaka et al. [10].

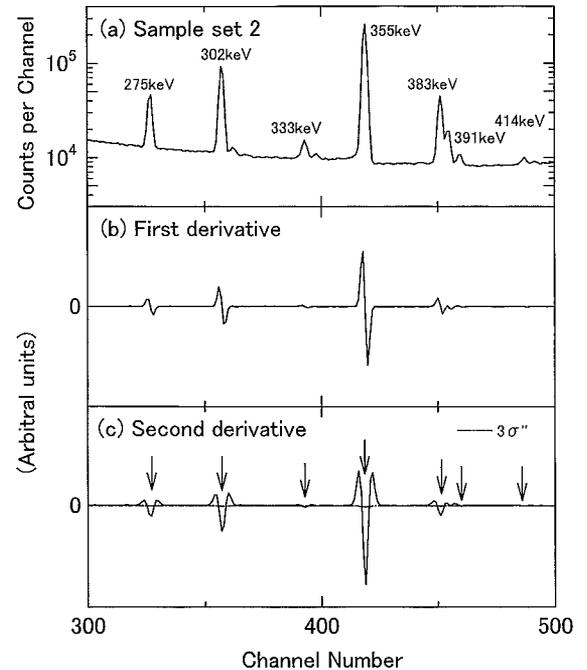


Fig. 3. An example of peak search procedure: (a) original gamma-ray spectra of sample set 2, (b) its first derivative, (c) second derivative. Peak position areas are indicated by arrows.

Table 2

Three sets of radioisotopes and output of ANN algorithm

| Sample set | Radioisotope | Output |
|------------|-------------------|--------|
| Set 1 | ^{54}Mn | 0.94 |
| | ^{60}Co | 0.99 |
| | ^{88}Y | 0.99 |
| | ^{137}Cs | 0.83 |
| Set 2 | ^{22}Na | 0.98 |
| | ^{88}Y | 0.99 |
| | ^{133}Ba | 0.95 |
| | ^{137}Cs | 0.97 |
| Set 3 | ^{22}Na | 0.93 |
| | ^{57}Co | 0.97 |
| | ^{88}Y | 0.99 |
| | ^{137}Cs | 0.95 |

Gamma-ray spectra from these ore samples are shown in Fig. 5. Sample 1 (SF84-1480) was taken from one of the reactor zones and sample 2 (SF84-1400) was from out of the reactor zone in the same

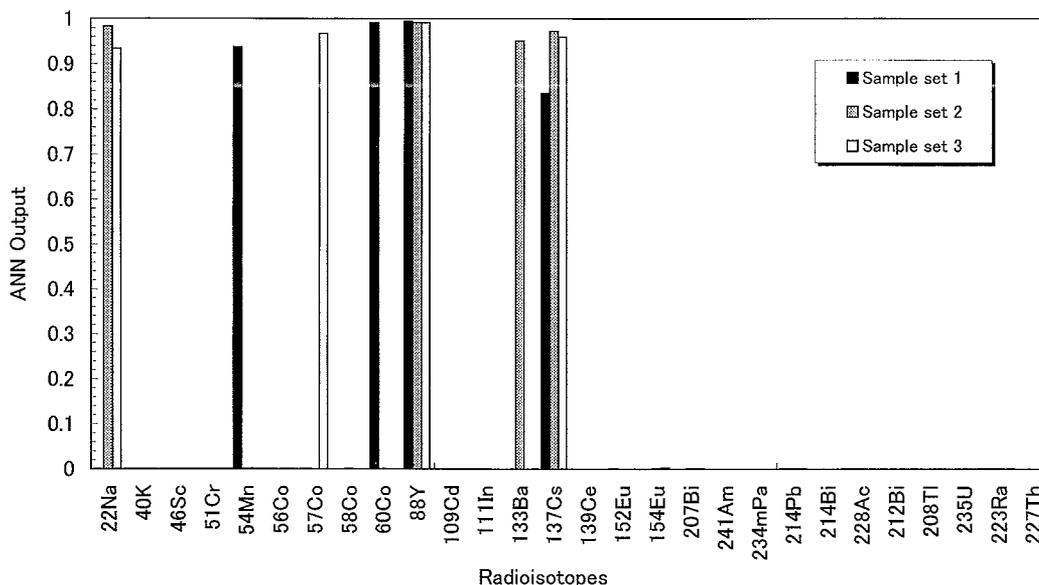


Fig. 4. Results of the ANN analysis for gamma-ray spectra from three sets of mixed radioisotopes.

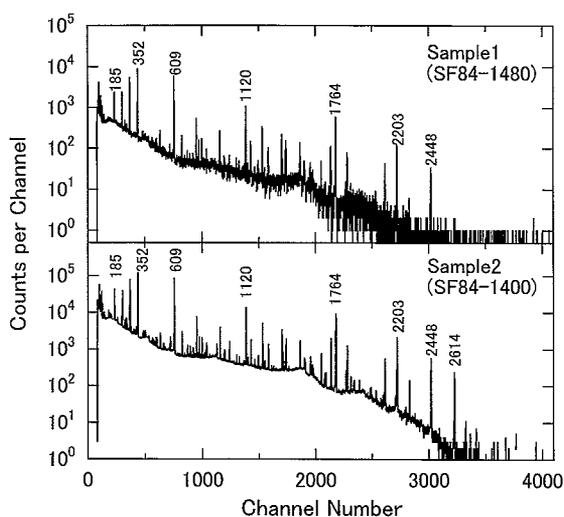


Fig. 5. Gamma-ray spectra of uranium ore samples obtained from the natural reactor zone at Oklo mine (Sample 1: reactor zone, Sample 2: out of reactor zone).

Table 3
Output of the ANN analysis for gamma-ray spectra of two uranium samples^a

| Radioisotope | Output | |
|------------------------|----------------------|----------------------|
| | Sample 1 (SF84-1480) | Sample 2 (SF84-1400) |
| <i>U-series</i> | | |
| ^{234m} Pa | 0.98 | 0.47 |
| ²¹⁴ Pb | 0.99 | 0.99 |
| ²¹⁴ Bi | 0.99 | 0.95 |
| <i>Th-series</i> | | |
| ²²⁸ Ac | 0 | 0.99 |
| ²¹² Bi | 0 | 0.99 |
| ²⁰⁸ Tl | 0 | 0.99 |
| <i>Actinium-series</i> | | |
| ²³⁵ U | 0.99 | 0.73 |
| ²²³ Ra | 0.92 | 0.21 |
| ²²⁷ Th | 0 | 0.29 |

^a Sample 1 (SF84-1480) is taken from the natural reactor zone and sample 2 (SF84-1400) is from out of the reactor zone.

core. These spectra, including more than 120 gamma-ray peaks, were processed to the present ANN and radioisotopes of uranium, thorium and actinium series were identified. Outputs of the ANN for two samples are given in Table 3, and

also depicted by the histogram in Fig. 6. It is clearly evident that radioisotopes of the uranium and actinium series are contained in both samples, but thorium-series radioisotopes are contained only in sample 2.

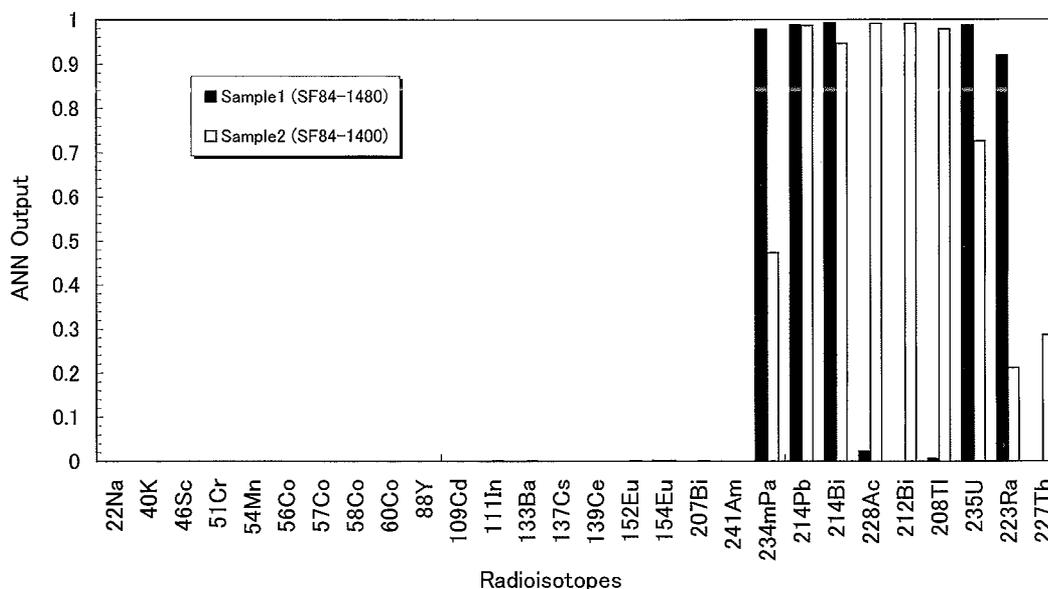


Fig. 6. Results of the ANN analysis for gamma-ray spectra of uranium ore samples.

Table 4

Comparison of peak intensity ratio between uranium samples from the natural reactor zone (SF84-1480) and out of the reactor zone (SF84-1400)

| Parent | Progeny | E (keV) | I_r (%) | Peak intensity ratio (N_{143}/N_E) ^a | |
|------------------|---------------------------|-----------|-----------|---|-----------|
| | | | | SF84-1480 | SF84-1400 |
| ^{235}U | | 143.7 | 10.5 | | |
| ^{238}U | ^{234}Th | 63.3 | 3.8 | 0.50 | 0.64 |
| | | 92.3 | 2.7 | | |
| | | 92.8 | 2.7 | | |
| | | 112.8 | 0.24 | | |
| | $^{234\text{m}}\text{Pa}$ | 1001.1 | 0.59 | 0.37 | 0.82 |
| | ^{214}Pb | 351.9 | 35.1 | 0.012 | 0.029 |

^a N_{143} and N_E indicate peak counts of gamma-ray energy 143 and E keV, respectively.

4. Discussion

It has been shown that the ANN algorithm can be usefully applied to the analysis of gamma-ray spectra by recognising the combination pattern of emitted gamma-ray energies. After radioisotope identification based on the present ANN, individual radioactivity can be determined from the peak counting rate corrected by the detection efficiency. The gamma-ray intensity balance must be also inspected during the calculation. In the rare situation where there is only a single gamma-

ray from a radioisotope and its energy overlaps with other gamma-rays, it becomes difficult for the ANN to identify the exact radioisotope, e.g., where ^{22}Na and ^{154}Eu have the same gamma-ray energy of 1274 keV. The existence of a single gamma-ray emitter can be found by examining the intensity balance of a multi-gamma emitter.

The ANN is especially superior in the analysis of gamma-ray spectra which contain numerous gamma rays such as uranium ores. The present ANN analysis of uranium ores indicated that radioisotopes of the uranium series were contained

in sample 1 and those of the uranium and thorium series were in sample 2. Mass spectrometric analysis [10] reported that $^{235}\text{U}/\text{U}$ is 0.005069 (7) for sample 1 and 0.007254 (15) for sample 2, respectively. A depletion of ^{235}U in samples from the reactor zone (sample 1) suggests that fission reactions have occurred at some time in the past [10]. The depletion of the $^{235}/^{238}\text{U}$ ratio in sample 1 is also indicated from the analysis of present gamma-ray spectra. The ratio of the gamma-ray peak counts of the 143 keV (^{235}U) peak were compared with those of the progenies of ^{238}U at 63, 113 keV (^{234}Th), 1001 keV ($^{234\text{m}}\text{Pa}$), 352 keV (^{214}Pb). The results are given in Table 4. It is indicated that the ratios of the 143 keV peak to the 113 and 352 keV peaks of the reactor zone sample are one-half of those of the reactor zone. The present results are consistent with the mass spectrometric analysis.

5. Conclusion

The ANN was applied to the analysis of gamma-ray spectra obtained by Ge detectors. To overcome the large input data size, a set of gamma-ray peak energies instead of raw spectral data was processed to the ANN. It has been shown that excellent pattern recognition of the ANN can

be usefully applied for radioisotope identification for gamma-ray spectra.

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