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Application of a neuro-fuzzy model for neutron activation analysis (NAA)

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Abstract. Neutron activation analysis (NAA) is a precise chemical multielemental method of analysis which is satisfactorily used for qualitative and quantitative analyses. Repeated irradiation is needed because of mal-determination of some elements due to peak overlap in qualitative analysis. In this study, NAA procedure has been modified using a neuro-fuzzy model to avoid repeated irradiation based on multilayer perceptrons network trained by the Levenberg-Marquardt algorithm. This method increases the precision of spectrum analysis in the case of strong background and peak overlap.

Key words: neural network • fuzzy logic • neutron activation analysis (NAA) • gamma-line energy

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Introduction

The NAA method is one of the most sensitive methods of isotopic analysis of samples. NAA relies on the (n, γ) activation of target nuclei by neutrons. In delayed measurement, the nuclear reaction produces radioactive nuclides in samples and the amount of radioactive atoms of each element are measured subsequently by gamma detectors [11]. Identification of the elements is based on the fact that most elements has characteristic gamma energies. Intensity of the emitted radiation depends on the number of atoms in the target sample so that quantitative detection can be performed by measurement of sample activity. With known parameters such as neutron flux, gamma-energy spectrum, cross sections and their energy dependence, irradiation time, half-lives and detector efficiency, the amount of elements can be calculated from the measured radioactivity. In practice, a reference (standard sample) is irradiated together with the original sample in the same irradiation conditions. The standard sample usually consists of a mixture of elements with known concentration of each element [1]. Two points in this method should be considered; a) qualitative and quantitative analyses are done separately and may require at least two irradiations for full determination and, b) gamma lines identification is very difficult in the presence of background and spectrum overlap, so that the probability of mal-determination of qualitative analysis may increase. To avoid these problems, a modification was applied in NAA using artificial neural network (ANN).

Neural networks are tools for non-linear data modeling. They can be used to model complex relationships between inputs and outputs or to find patterns in data [5]. One advantage of the neural network approach

is that most of the intense computation takes place during the training process before analysis [6].

ANN is used in a wide variety of processing applications where a real time, data analysis and information extraction are required. Once the ANN had been already trained for a particular task, operation was relatively fast and unknown samples could be rapidly identified. In this work library the least squares (LLS) method accomplished with a neuro-fuzzy model is used for this purpose. It is based on separation of the measured gamma spectrum, on components characteristic of spectra of isotopes-subject of research. This component requires availability of the library of reference spectra. In this work, this library is produced by a combination of some experimental respect, Monte Carlo (MC) simulation and the neuro-fuzzy model. The LLS approach for NAA has a number of advantages [3] such as: 1) It is the most fundamental approach; 2) It is capable of providing accurate results since the entire spectrum can be used; 3) It automatically provides an estimation of the standard deviation of each calculated elemental amount in the presence of all other components; 4) Peak interferences are automatically accounted for.

The only requirement is that elemental libraries must be available for all elemental samples. In this study, it is necessary to have a library for all elements. This library was produced by neural network by considering a combination of some experimental data and the MC simulation.

Material and methods

In NAA, the quantity of an element m_{smp} is given by the following equation:

$$m_{smp} = m_{std} \times p_{csmp}/p_{cstd} \quad (1)$$

where: m_{std}, m_{smp} are the standard and sample masses, respectively; p_{csmp}, p_{cstd} are the net area under gamma peak in the comparator and sample, respectively. Interval time between the end of irradiation and the start of spectroscopy can be accounted for correction related to gamma energies according to Eq. (2):

$$p_c = p / e^{-\lambda T_d}(1-e^{-\lambda T_c}) \quad (2)$$

where T_d is the interval time between the end of irradiation and the start of spectroscopy; T_c is the time of counting in spectroscopy; p and p_c are the uncorrected and corrected net area under gamma line, respectively. Some elements have more than one gamma line, so the product of detector efficiency and the ratio abundance of gamma line of the element can be considered as the criterion for each element [4]. In other words, the gamma line with the largest value of the criterion can be participated in element identification. Noting the linear nature of the measurement, the largest intensity of each element multiplied by the counts in each channel (or pulse-height energy bin per unit element) is proportional to the elemental weight ratio. Moreover, the count in each channel is equal to the sum of counts for each element. For any sample with unknown mixture, the intensity is considered as a sum of all elements in each channel (in a least-squares sense). For each channel i (or energy bin), this is mathematically stated below:

$$y_i = \sum_j^m x_j a_{ij} + E_i, \quad i = 1, n \quad (3)$$

where: y_i are the counts of the sample with unknown mixture in the channel i; x_j is the gamma spectrum of the element j in the sample; a_{ij} are the counts in the channel i of element j in the sample with unknown mixture; E_i is the random error in counts in the channel i; m is the total number of chemical elements constituting the sample and n is the total number of the multichannel analyzer (MCA) energy channels. The reduced chi-square value (χ_v^2) is formed according to the following relation:

$$\chi_v^2 = \frac{1}{n-m}\sum_{i=1}^{n} E_i^2 / \sigma_i^2 \quad (4)$$

where σ_i^2 is the variance of y_i and is usually taken from Poisson distribution and, therefore, equal to y_i. The x_j is found by minimizing the reduced chi-square value. This is done in the usual way by setting the derivation of the reduced chi-square value with respect to each x_j equal to zero to obtain a system of m equations. These equations can be solved simultaneously by the inverse matrix method to find x_j. Then, qualitative and quantitative analysis of trace elements is a proper normalization coefficient (a_{ij}) in Eq. (3).

A neuro-fuzzy model is modified to determine normalization of a_{ij} by minimizing the error of the approximated spectrum. One feature of this technique is that it uses the whole spectrum in the identification process instead of only the individual photo-peaks. For this reason, it is potentially more useful for processing data from lower resolution gamma-ray spectrometers. This approach has been successfully tested with data generated by MC simulations and with field data from both sodium iodide and germanium detectors [8].

The following steps should be considered to modify quantitative NAA using neuro-fuzzy model as shown in Fig. 1:

1. Collection of experimental data (experimental or simulation results).
2. Proper neural network model selection.
3. Training of recommended neural model.
4. Evaluation of trained neural model.

In the first step, data should be obtained from experimental or MC simulation processes for NAA. For experimental data, the collection (data acquisition) in NAA, a 60 cm^3 P-type high purity germanium (HPGe)

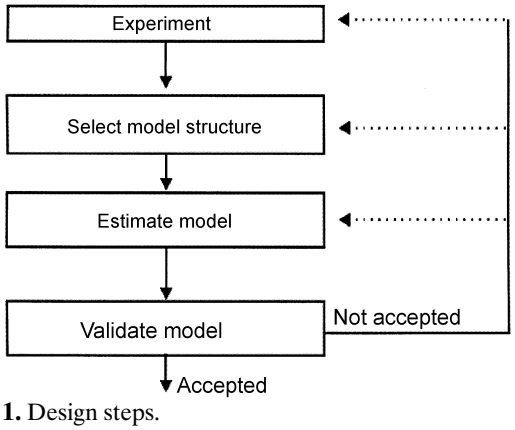

Fig. 1. Design steps.

Table 1. ANN structure parameters

Multilayer perceptron	Type
1024-1024-63	Architecture
Log sigmoid	Activation function

detector with a resolution of 0.75 keV full width at half maximum (FWHM) at a photon energy of 122 keV and of 1.8 keV FWHM at 1332 keV was used. After irradiation, activity of the sample can be determined from experimental conditions such as irradiation and cooling time, neutron flux and irradiation position as well as elemental nuclear characteristic [12].

All of the effective parameters in experimental conditions to form a gamma peak have been fixed, so that irradiation time is the only variable factor to predict the quantity of element. These conditions were simulated by MCNP-4C code (Monte Carlo N-particle transport code) [2] to complete training data procedure, so that the gamma spectrum was obtained from different samples in various conditions. Gaussian broadening of gamma peaks was calculated from the least squares method by determining detector efficiency. MCNP outputs from related tallies are considered similar to MCA for exact comparison. Structure of the elemental composition was considered in such a way that all of the elements could be identified at least in one of the spectra. The required data prepared for training of neuro-fuzzy model uses preprocessing such as omitting of the single escape, double escape and sum gamma peaks as well as normalized spectrum.

The selection of appropriate structure for neural network model is a subsequent phase. The prototyped ANN was constructed as a multilayer perceptron network and was trained with the Levenberg-Marquardt algorithm by using a training set from the collected data. The parameters used to train this ANN are listed in Table 1. Neurons in the input layer, meaning the energy bin of gamma from 100 keV to 3.6 MeV and output layers neuron, show the concentrations of elements in the unknown sample.

Figure 2 shows the two experimental trained spectra. Figure 2a shows the gamma spectra from an experimental sample containing 3 mg of ^{38}Cl and 4.8 mg of ^{193}Ir. Figure 2b shows the gamma spectra of the experimental sample containing 19.97 mg of ^{71}Zn.

Neural network can identify elements by comparing differences and similarities in the trained spectrum and then making an elemental spectrum library by weight adjusting. After these steps (training neural network and making elemental spectrum), the LLS method can be applied. Gamma counts in energy bins and concentration of elements is introduced as an input and output of the model, respectively. After the training step, evaluation and validation of the model should be carried out. For this purpose, the residuals (prediction errors) of test samples are used.

The collected data are divided into two sets, one is used for model training and the other is used for model validation. Different experimental samples were analyzed by this model showing a good accuracy of the neuro-fuzzy model. To minimize probable errors of the model in element diagnosis (with more than one gamma peak for identification) the detector efficiency

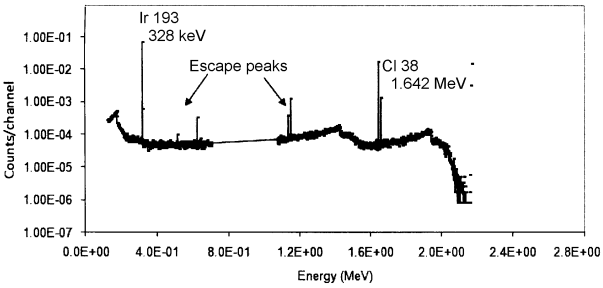

Fig. 2a. A sample of learning data containing ^{38}Cl and ^{193}Ir.

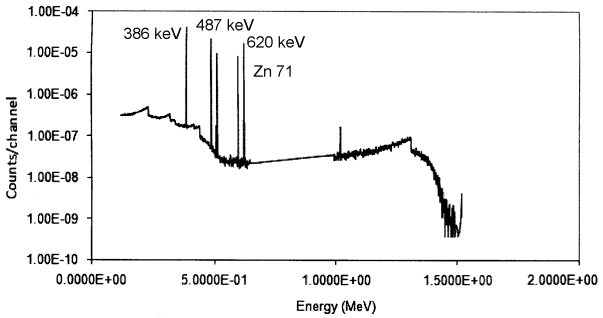

Fig. 2b. A sample of learning data containing ^{71}Zn.

and gamma-line abundance ratio and their product were used. In other words, the necessary condition for the existence of an element in the spectrum is the presence of gamma line with a maximum abundance. The next condition is the half-life of radioisotopes. For short term irradiation, long half-life radioisotopes were not considered and *vice versa*. After these considerations, the list of recommended elements in the sample was restricted. Qualitative and quantitative analyses should be independent of irradiation parameters and the neural model should show the same results for various irradiation times. One method is the use of neural model training for all probable irradiation times, but for its time consuming and massive output data production, this method is not practical for use. Another method is the training of some neural model for specific irradiation times and using a fuzzy system to interpolate for all the irradiation

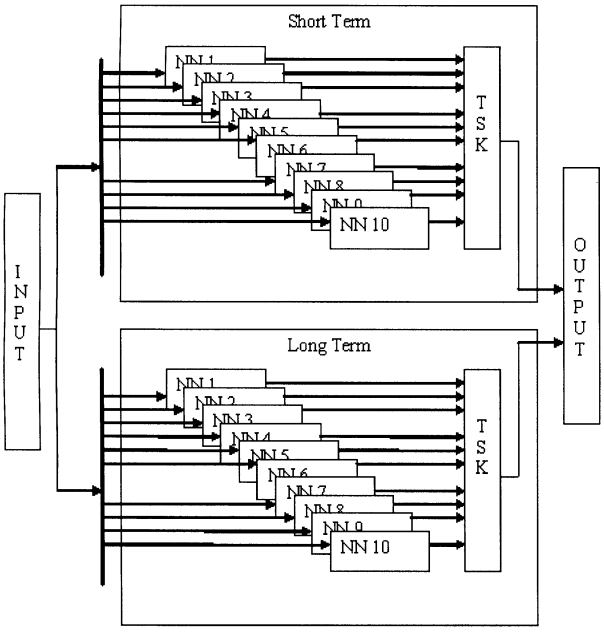

Fig. 3. Neuro-fuzzy model.

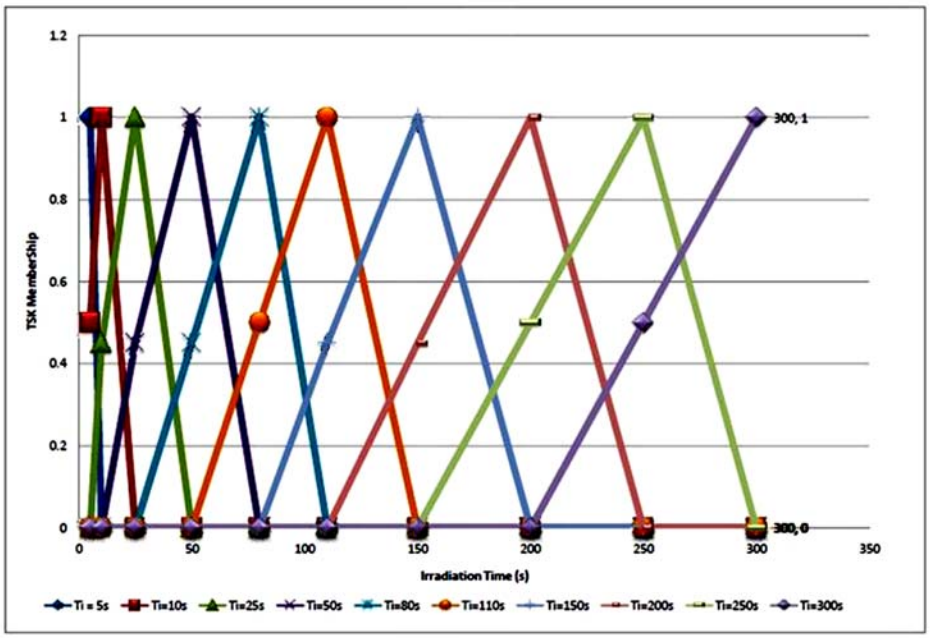

Fig. 4. TSK membership functions for short-term group.

times. Two groups of neural models were considered for short and long-term samples regarding half-lives. Each of them includes 10 subcommon models according to Fig. 3. Irradiation times for short-term samples are considered as 5, 10, 25, 50, 80, 110, 150, 200, 250, 300 s and for long term are 5, 15, 35, 65, 105, 160, 220, 290, 370, 480 h. Neural model is trained for all of the above irradiation times. The Takagi-Sugeno-Kang (TSK) fuzzy system [10] is used to interpolate between these times. The rule of fuzzy system is:

Rule i: if $T_{\text{irradiation}}$ is T_i then out = NN_i (x, $T_{\text{irradiation}}$)

T_i = 5, 10, 25, 50, 80, 110, 150, 200, 250, 300 s – for short term,

T_i = 5, 15, 35, 65, 105, 160, 220, 290, 370, 480 h – for long term.

TSK membership functions have been chosen in such a way that in a desired irradiation time, two fuzzy rules or neural models are activated and a fuzzy system produces a favorite output from combining these activated outputs. Figure 4 shows the TSK membership functions for short-term group.

Results and discussion

The neuro-fuzzy model illustrated in Figs. 5 and 6 was tested by standard sample, SL-1, with known elemental concentrations. 10 mg of this sample was irradiated for 1 min and then cooled for 35 s. Its spectrum was recorded for 5 min under laboratory conditions and it is shown in Fig. 5 [9]. Figure 6 shows the recorded gamma spectrum of this sample irradiated for 5 h. The SL-1 sample elements are listed in Table 2 [7].

As shown in Figs. 5 and 6, the spectrum contains short and long-lived gamma peaks, double and single escape as well as sum peaks. Short-term and long-term components of gamma spectrum of the sample were introduced as an unknown input into the neuro-fuzzy model. Mass of elements and uncertainty of elemental analysis performed by NAA were reported in Table 3. These measurements were performed through several experimental analyses in the same conditions. Elements

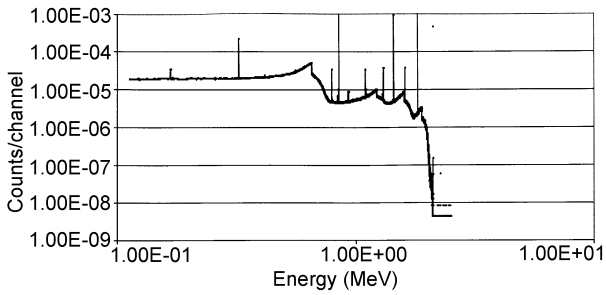

Fig. 5. Gamma spectrum of SL-1 in short-term irradiation (1 min).

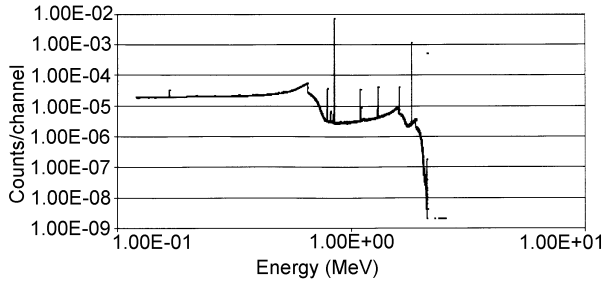

Fig. 6. Gamma spectrum of SL-1 in long-term irradiation (5 h).

Table 2. Mass of elements in 10 mg of certified reference material SL-1 (weight (mg) ± uncertainty (% of weight))

Element	Weight (mg)
Iron (Fe)	0.674 ± 0.17%
Manganese (Mn)	0.0346 ± 0.016%
Titanium (Ti)	0.0517 ± 0.037%
Arsenic (As)	0.000275 ± 10.5%
Barium (Ba)	0.00639 ± 8.29%
Cerium (Ce)	0.00117 ± 14.5%
Cobalt (Co)	0.000198 ± 7.57%
Rubidium (Rb)	0.00113 ± 9.73%
Thorium (Th)	0.00014 ± 7.14%
Vanadium (V)	0.0017 ± 8.82%
Zinc (Zn)	0.00223 ± 4.48%

Table 3. List of determined elements

Element	NAA in the laboratory (mg)	Neuro-fuzzy (mg)
Vanadium (V)	0.00170 ± 0.000034	0.00182 ± 0.000058
Rubidium (Rb)	0.00113 ± 0.000056	0.00109 ± 0.000023
Manganese (Mn)	0.0346 ± 0.0014	0.0349 ± 0.00012
Barium (Ba)	0.00639 ± 0.00025	0.00642 ± 0.00011
Titanium (Ti)	0.0517 ± 0.0021	0.0506 ± 0.0019
Arsenic (As)	0.000275 ± 0.000011	0.000298 ± 0.0000079
Iron (Fe)	0.674 ± 0.013	0.625 ± 0.023
Lanthanum (La)	0.00052 ± 0.0000052	0.00056 ± 0.000016

revealed by the neuro-fuzzy model along with their weights are also listed in Table 3. As seen, the error of weight estimation is under 10%. In a typical NAA software, for each gamma line, more than one element may be assigned and it is up to users, according to their experience, which element is the correct one; that is why this method may have a low accuracy and takes a long time to analyze spectra. The present model can save time as well as enables precise determination of qualitative and quantitative NAA. It has the capability of identifying elements in the spectrum even with noisy or imperfect data that can be induced from high background or bad calibration of spectroscopy equipment.

Conclusion

In this study, an intelligent model was introduced using a neural network and a fuzzy system to do precise qualitative and quantitative NAA in one stage irradiation. This model has used the multilayer perceptron (MLP) neural network and TSK fuzzy system for function approximation and parameter estimation of LLS method. A standard sample was used for evaluation and validation of the model and irradiated as an unknown sample and its spectra was introduced as an input to the model. One of its advantages is a high precision of anticipation even by imperfect or noisy input data. By using this method, the peak overlap is decreased and there is no need to do repeated irradiations. Output results show a satisfactory agreement regarding the speed and accuracy of qualitative and quantitative NAA.

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