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Artificial Neural Network: A tool for Rapid Quantitative Elemental Analysis Using Neutron Activation Analysis

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ABSTRACT: An artificial neural network (ANN) has been trained with real-sample NAA (neutron activation analysis) spectra of irradiated materials. Following the training stage ANN was applied to a subset of similar samples thus obtaining the elemental concentrations by NAA method. Results obtained with the ANN method are in good agreement with results obtained from NAA techniques, showing the high potentiality of ANN in NAA quantitative analyses.

KEYWORDS: Artificial Neural Network, Neutron.

I. INTRODUCTION

Developments from artificial intelligence like artificial neural networks, ANN, can be regarded as an engineering procedure emulating the human brain activity. It can be defined as a set of non-linear and non-stationary interconnection of elemental processes able to carry out at least one of the following functions: training, remembrance and generalization, or, abstraction of substantial properties. The fundamentals of the ANN technique have been described elsewhere [1- 2] and several commercial and free codes are accessible nowadays. Applications of ANN to atomic and nuclear physics has increased during the last two decades [3–13], mainly in problems related to nuclear reactors. The most used paradigms in artificial intelligence applications to nuclear science and particle physics are the expert system, general algorithms, fuzzy system, neural networks and hybrid system. Some applications to α , β , and γ spectra have been reported in the last decade. Further application of ANN in other cases where strong non-linear effects are present like in the spectral analyses generated in analytical techniques like PIXE and XRF (X-ray fluorescence) are scarce and in BIXE spectra (beta induced X-ray emission) remain unexplored [14,21].

Application of ANN is found in several issues where similar patterns have been represented by many spectra with specific differences. A typical case is presented in the elemental analysis of samples of unknown composition by neutron activation analysis (NAA) technique. The corresponding spectra represent the peaks containing known energy with variable intensities holding necessary information for the evaluation of the primary concentrations present in each sample. NAA is a nuclear technique based on the detection of γ -rays emitted by radionuclides neutrons [2–5]. Activation analysis differs from the other methods such as chemical analysis; it depends on the properties of the nucleus and not on the behavior of electrons in the outer shells of the atom. In general, NAA has many advantages over many conventional methods such as simultaneous analysis of multi-component samples and high degree of sensitivity for the majority of elements, many elements can be determined at levels of parts per billion. The basic essentials required to carry out analysis of samples by NAA are: source of neutrons, suitable instrumentation for detecting gamma rays, and a detailed knowledge of the reactions that occur when neutrons interact with target nuclei. One of the factors that define the quality of the analysis is the ability to determine the position of peaks in the spectrum, calculate the energy of the gamma ray each peak represents and estimate the areas of the peaks in the spectrum. Usually this is accomplished by the use of some reliable computer code, like Genie 2000 software (Canberra Industries, Meriden, USA). Additional inputs of the proper physical parameters involved in the irradiation like beam flux, detector efficiency, reaction cross section geometrical parameters, etc., permit the determination of the elemental concentrations.

The purpose of this work has been to take advantage of the non-linear properties and the neural network ability to learn and generalize together with its fast response. Samples of cement were irradiated with neutrons to determine their

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elemental concentrations as part of a study of determining trace elements responsible of long lived isotopes in cement used reactor shielding. Several gamma spectra were obtained from cement samples which apart of the samples set was analyzed by the standard method of NAA and later used to train the ANN for further calculations. The proposed ANN method was applied for quantitative analysis of irradiated cement samples.

II. ANALYSIS

Principle of NAA

There are three known methods for determining elemental concentrations by NAA namely: absolute, relative, and comparator methods [22]. We use in this work the absolute method because it offers several advantages on the basis of speed, cheapness, versatility and ease of automation. It also offers the possibility of multi-element analysis in one single irradiation. Consider a sample of mass W , subjected to irradiation in a neutron flux for a time period (t_i). At the end of the irradiation the sample is allowed to decay for a time t_d and counted for a time t_c . If the mass of an element in the sample is m , then, the measured, c can be obtained from the basic absolute neutron:

$$C = m / w = \frac{P}{w [N_0 / M] \Omega \sigma \Phi \gamma \epsilon S D C t_c} \quad (1)$$

where P is the net peak area under the gamma peak of interest, M is the atomic mass of the target nuclide, N_A is Avogadro's number, Ω is the natural abundance of the target isotope, σ is the effective cross section for (n,γ) reaction, γ is the gamma-ray emission probability ϵ is the photo peak efficiency, S is the saturation factor, $S = (1 - e^{-\lambda t_i})$, D is the decay factor, $D = e^{-\lambda t_d}$ and C is the counting factor $C = (1 - e^{-\lambda t_c})$ where λ is the decay constant for the (n,γ) product radionuclide.

Principle of ANN

As mentioned above, the net peak area under the gamma peak of interest is currently used to determine the elemental concentration. Here, a slightly different approach has been used based on the fact that ANN application disregards additive constants. Then considering the observation that the background contribution at the peak is rather constant for equal irradiation times, the use of the maximum value at the peak channel as an input in ANN calculations, simplifies data handling and provides equivalent results as those obtained when the full area is used. These values and the already known concentrations were given as an input and output data respectively in the ANN training phase. fig. 1 shows the diagram followed in each phase.

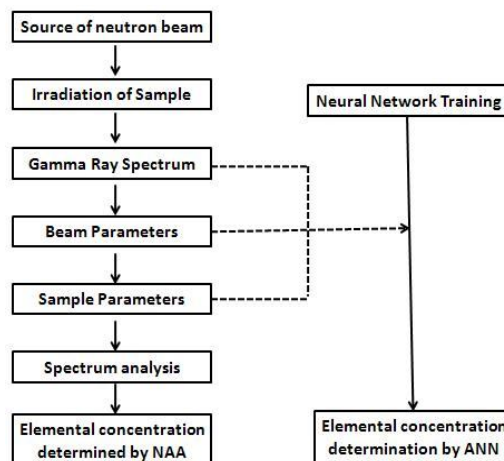


Figure 1: A diagram showing the relationship between ANN inputs and the classical NAA experimental steps. A feed-forward Artificial Neural Network (ANN) has been applied in the training phase right on the concentration data

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secured from real time analysis. The network was trained with the well-established back-propagation algorithm in order to learn first and then reproduce the process in order to obtain the concentration of certain elements in some cement samples. The neural network feeds with input parameters like: the neutron beam flux impinging the sample, mass of the sample, spectral information of the detected γ -ray.

For each element, an ANN with three layers (fig. 2), (input, hidden, output) was used to generate the mapping of the p -input parameters (in this work $p = 4$ to the q -output parameters, here represented by the concentration of the element in the sample ($q = 1$)). The mapping was conducted considering the weights of the respective neuron connections which were recalculated during the supervised training procedure.

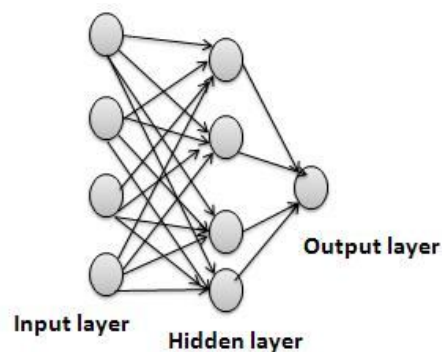


Figure 2: Schematic representation of a three-layer feed-forward neural network.

For training, the method for least square error back propagation, the most often used supervised learning algorithm, was used. Supervised learning refers to the known input and expected output for every example in consideration; therefore, training the ANN considers the fact that the calculated outputs should be in vicinity to the expected outputs strictly for every considered data points. Apparently, a neural network considering with p -inputs, single hidden layer and with L -neurons (here considering $L = 4$) and a single output node, The resultant error rate E_k for the input or output training scheme (x^k, y^k) is represented by

$$E = \frac{1}{2} \sum_k (y^k - o^k)^2 \quad (3)$$

where o^k is the resultant calculated output produced while the k -th input vector $(x_1^k \dots x_p^k)$ is been made obtainable to the network and sigmoid function is used for the transformation and where y^k represents the expected output value in relation to input x^k . The altogether computation of the error is done by

$$E = E_k \quad (4)$$

The protocol for altering weights considering their presentation of input or output pair k is performed by the gradient descent method, suggesting the minimization of the quadratic error function in an iterated manner. Once the training was successfully completed, effective testing for verifying the efficiency was performed. To train the ANN, first the initial weights are fixed in a random way. Then each sample (the 4 input values) is introduced in the ANN, which compute the output (O_k in the above equations), it is say, the actual concentration value. This value is compared with expected concentration value and according to the error, the weights are modified so that both values (computed concentration and expected concentration) become the closest possible.

In general, defining the training set is performed experimentally. In the present study, due to the lack of sufficient available samples and following the experience of authors training set of 5 spectra was selected randomly and a testing set of 3 spectra were considered for each ANN. Results confirmed that this selection was adequate.

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Training of the ANN considering each of the elements and similar topology was done in an independent manner where sigmoid activation function was implemented for the hidden and output layers. Error value, momentum, learning rate, etc., parameters and the topology were obtained through laborious experimentation.

III. RESULTS AND DISCUSSION

Without missing generality in this ANN application only the major elements observed in the spectra were considered. Trace elements, although important for other studies, were not statistically useful for the present purpose. Elemental concentrations were calculated by standard NAA methodology in the way described above. These results were considered as „„expected concentrations““. As for NAA uncertainties, overall values ranged between 3% and 12%. A feed-forward neural network, with definite topology was independently trained in parallel with the backward error propagator to determine the elemental concentrations of interest in the samples of this study. Spectra were randomly selected and used in training the network and three were used in testing it. All training cases were learned correctly by each of the six neural networks. The agreement is illustrated in Fig. 3, where concentrations are shown for each element in samples. To assess the goodness of the ANN method in providing concentrations values, a comparison with NAA results was done and is shown in fig. 4. A linear regression calculation for results from all elements, in the test and training sets, was performed. Correlations factor, r^2 , of 0.86 and 0.99 were obtained for the test and training sets, respectively.

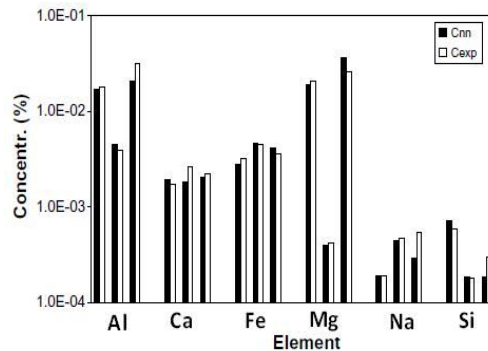


Figure 3: Elemental concentration obtained from the three samples of the test set using ANN (C_{NN}) and NAA (C_{exp}), are shown for three cement samples respectively for each element.

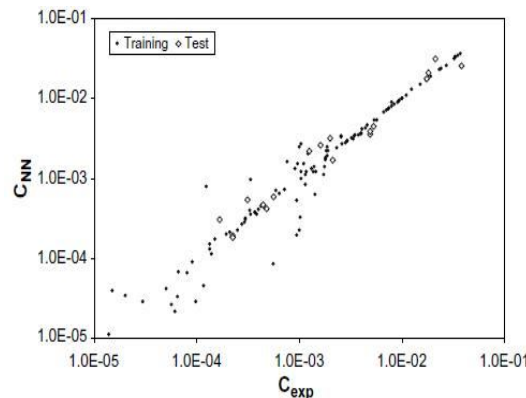


Figure 4: The predicted concentrations from ANN (C_{NN}) as compared with NAA (C_{exp}) concentrations.



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IV.CONCLUSIONS

The main purpose of this work was to explore the application of ANN for a fast analysis of NAA in a study of elemental concentration of some cement samples. It has been proved that this methodology provides results as good as those obtained by one of the already accepted methods. Even though the number of cases was rather limited, the decision to use independent neuronal topologies for each element working in parallel provided reliable results. Surely, the experimental results would be improved with a greater number of cases to use as training and test sets. So, the use of ANN as a reasonable alternative for the elemental concentration determination in NAA procedure has been established. In addition, this ANN application proved to be fast enough as to predict that the analysis of a great number of samples can be accomplished in few minutes, avoiding the direct participation of the analyst in each one. Once the ANN has been trained, its use becomes fast, thus, permitting the automation of NAA spectra analysis and reducing costs and time.

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