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Neural Network Approach Coupled with MCNP to Analyze Gamma Spectra for Bio-samples

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Abstract – *While Neutron Activation Analysis is widely used in many areas, sensitivity of the analysis depends on how the analysis was conducted. Even though the sensitivity of the techniques carries error, compared to chemical analysis, its range is in parts per million or sometimes billion. Due to this sensitivity, the use of neutron activation analysis becomes important when analyzing bio-samples. Neural Network is an attractive technique for automated systems. Although there are Neural Network applications on spectral analysis, training by simulated data and analyzing experimental data has not been studied. This study offers an improvement on gamma spectrum analysis and optimization on Neural Network for the purpose. The current work considers five elements which are considered as trace elements for bio-samples. However, the system is not limited to bio-sample analysis. The only limitation of the study comes from data library available on MCNP. Better results were obtained when Neural Fitting tool in MATLAB was used. As a training function, Levenberg-Marquardt algorithm was used with 23 neurons in hidden layer with 260 gamma spectra in the input. Since the interest of the study deals with five elements, five neurons representing peak counts of five isotopes in the input layer were used. Five outputs neurons revealed mass information of these elements in irradiated Kidney stones. Results showing max error of 17.9% in APA, 24.9% in UA, 28.2% in COM, 27.9% in STRU showed the success of Neural Network Approach in analyzing gamma spectra. The simulation and experiments were made under certain experimental setup (3 hours irradiation, 96 hours decay time, 8 hours counting time). Yet, the approach is subject to be generalized for different setups.*

I. INTRODUCTION

Neutron activation analysis (NAA) is a very sensitive technique to identify and determine concentration of elements in a sample [1, 2]. NAA here, refers to delayed gamma spectra analysis which consists of irradiating a target and transferring it to a detector. Then, Gamma spectra give information about element identification and quantification. NAA may become complex especially when dealing with bio-samples [2, 3]. In human organisms, some of heavy elements are used as inhibitors. More than adequate amount makes them toxic for living organisms [2]. Thus, accurate quantification of these elements is crucial for medical science.

Concentration of elements, relative to one chosen element, can be obtained from gamma spectra by peak analysis. This technique consists of comparing two different spectra from different samples under same experimental conditions.

$$m = \frac{P_k A_i \lambda_{i+1}}{\epsilon(E_k) e_k a_i N_A \sigma_i \phi (1-e^{-\lambda_{i+1} t_0})(e^{-\lambda_{i+1} t_1} - e^{-\lambda_{i+1} t_2})} \qquad (1)$$

where P_k is the net counts under the peak at energy E_k, λ_{i+1} is the decay constant of isotope with atomic number A_{i+1}, N_A is Avogadro's number, σ_i is the neutron absorption cross-section of isotope with mass number A_i, ϕ is the neutron flux, $\epsilon(E_k)$ is the detector efficiency at E_k, t_0 is irradiation time, t_2-t_1 is the counting time, e_k is the emission probability of the decay, and a_i is natural abundance of isotope A_i [4].

The advantage of this analysis occurs comparing same elements in different samples. In such a case, most of the terms in Equation (1) are canceled out. Only peak counts and mass of samples remain. When different elements are compared, there are some terms that propagate error through the analysis. Details of this approach can be found in Reference [5]. Furthermore, comparator technique which is a comparison of known and unknown samples can also be employed. Irradiating known and unknown samples under same experimental setups allows finding true concentration with a very small associated uncertainty. Detector dead time, uncertainty in reaction cross-sections, radiation yield of the detector material, detector resolutions, etc. have important effects in the analysis [2].

Machine Learning is an attractive technique to simplify the analysis process. Neural Network (NN) is one of Machine Learning classes that recognize behavior of a system with data carrying noise [6]. NN can be defined as a mathematical representation of how human brain works. It learns the behavior of a system from pre-existing inputs and outputs. A simple representation of a network consists of an input layer which receives data, one or more hidden layers and an output layer. Hidden layers play crucial role finding weights in between connected neurons by deciding whether to pass a signal to output layer. A single neuron is the fundamental unit of NN layers. Like a human neuron, it receives signals from incoming connections to be weighted, summed and passed through a threshold. Repetition of this process with many neurons in the layers trains the NN for correct weights between connections. Once the NN is

trained, it can be used to analyze an unknown set of data [7, 8].

In NN analysis of a system, the time consumption occurs only during the training process. With a trained NN, it does not take much time to analyze the system even though the system is very complex [7, 8].

NN has been used in many areas including engineering, science, medicine, business, etc. Advancement of computer technology as well as NN algorithms attracts more researches recently [7]. It has been applied in nuclear engineering field including NAA. Quantitative prediction of compounds, mixtures or elements [9-16] and peak identification [17-20] were studied with variety of methods. Yet, the current techniques are still under improvements.

NN is generally used for clustering, data fitting, pattern recognition, optimization, control, prediction with several training algorithms. Famous algorithms are Levenberg-Marquardt (LM) backpropagation, Bayesian Regularization (BR) backpropagation, Scaled Conjugate Gradient (SCG) backpropagation.

LM algorithm can be simply defined as a modified Gauss-Newton method. Equation (2) gives mathematical representation of LM algorithm.

$$x_{k+1} = x_k - [\boldsymbol{J}^T(x_k)\boldsymbol{J}(x_k) + \mu_k \boldsymbol{I}]^{-1}\boldsymbol{J}^T(x_k)\boldsymbol{v}(x_k) \qquad (2)$$

J, μ_k are Jacobian matrix and increment in eigenvalue of Hessian matrix (**H**=$\mathbf{J}^T\mathbf{J}$) respectively. υ(x) is error function that its sum squares are the performance index. Performance index is the function that NN algorithm minimizes by adjusting the weights. If μ_k is decreased to zero, the algorithm becomes Gauss-Newton approach [7]. Simply this algorithm is a numerical approach.

BR uses Bayes' Theorem for minimization. According to the theorem; if there are two random events (A and B), conditional probability that one event occurs represented with Equation (3).

$$P(A\backslash B) = \frac{P(B\backslash A)P(A)}{P(B)} \qquad (3)$$

BR approach in NN assumes that weights between neurons are random. They are assumed to be a Gaussian distribution. For very big size of training data, BR and LM result same error since one approach is numerical and the other is probabilistic [7].

SCG is efficient for large networks for pattern recognition. This algorithm is a modified version of conjugate gradient which works only for positive definite value of Hessian function [21]. Equation (4) represent estimate term s_n for non-zero input quantity$x_n = \tilde{p}_n^T s_n$.

$$\boldsymbol{s}_n = \frac{E'(\tilde{w}_n + \sigma_n \tilde{p}_n) - E'(\tilde{w}_n)}{\sigma_n} + \lambda_n \tilde{p}_n \qquad (4)$$

p is non-zero weight vectors, E(w) is error function, λ is adjusted scaler in each iteration to find estimate term, σ can be a positive value very close to zero [21].

LM algorithm requires more computer memory and takes shorter time during training while BR algorithm results the opposite. SCG algorithm training stops in short time. However, the stopping criteria for training depends on improvement of the mean square error that is also system dependent [22]. While MATLAB uses mean square error in the NN fitting application, an outside validation is necessary to optimize the application. Equation (5) represents relative error to analyze how good the system is.

$$Error\ \% = \frac{|X_{Experimental} - X_{Simulated}|}{X_{Experimental}} x100 \qquad (5)$$

Furthermore, the main success of an NN depends highly on training data set. Obtaining a big set of delayed gamma spectra is a concern especially when looking for trace elements in bio-samples because it may require longer irradiation and measurement time [5]. Collection of this type of data may take years depending on availability of the research reactor and equipment at Missouri University of Science and Technology.

MCNP [23] is a Monte Carlo simulation code for particle transport, which is useful in generating the required data set. However, available nuclear library is a limiting factor for delayed gamma production [24].

NAA has been performed via NN tool with MATLAB. The training data set was generated with MCNP to analyze experimental spectra. In referenced studies, NNs were trained with either simulated data to analyze simulated data or experimental data to analyze experimental data. The disadvantage of former is inapplicable to real problems while the disadvantage of latter is difficulty of required size of data. In the current research, adequate data set was generated with MCNP. Despite the fact that NN is useful, there is no solid guide for NN application on NAA. Thus, an optimization of NN was carried out as well. The focus of this study is on trace elements primarily (Gold, Bromine, Potassium, Zinc, and Sodium) in bio-samples. The choice was to compare results the previous study. However, the procedure can be easily generalized for different elements and compounds.

Primary idea of this work is to train an NN with simulated data and analyze experimental spectra for element concentrations. Monte Carlo simulation was employed to sufficient data set to train the NN.

II. DESCRIPTION OF THE ACTUAL WORK

The procedure is divided into several steps as shown in Fig. 1. Experimental data and simulated data were obtained. A calibration has been carried out to match MCNP output with known sample. Generated data set was used to train the NN to analyze an unknown sample. Four type kidney stones

were used with a known concentration to be treated as unknown sample to validate NN system. NaCl sample with known amount was used for calibration and traditional analysis of kidney stone samples to compare the NN results.

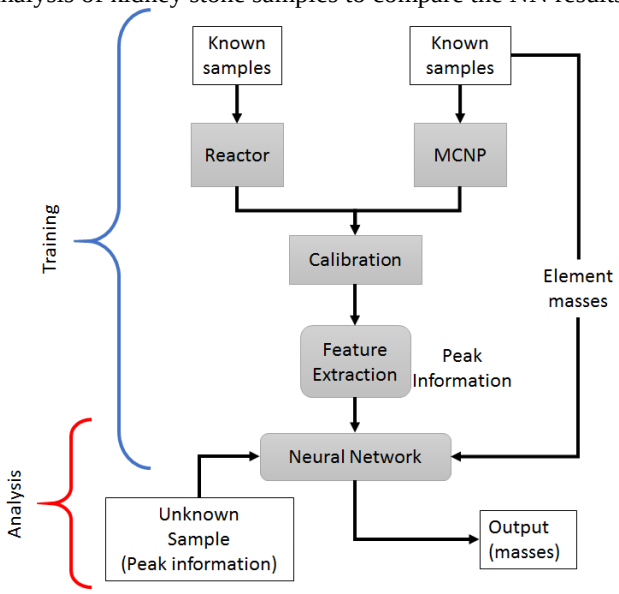

Fig. 1. Flow chart of procedure.

1. Experimental Data

Kidney stone samples with unknown compositions were irradiated for 3 hours at the Missouri S&T Reactor (MSTR). After decay time of 96 hours, experimental gamma spectra were obtained from high purity Germanium (HPGe) detector (Canberra BE3825). Details for the procedure can be examined through reference [5]. Under the same experimental setup, 5 mg salt (NaCl) sample was measured after proper preparation. The sample was dried because of humid. And, it was measured in a closed and sealed container. 5 mg salt contains 1.9669 mg Sodium. Gamma peak from Sodium carries information related to this mass. The purpose for irradiating Na is to evaluate and determine correction values for the simulation. Another advantage of Na in validation is that ^{24}Na has emission at 1.369 MeV 100% of time. Thus, comparison from decay to number of photons can be calculated easily.

2. Simulated Data

Monte Carlo simulation using MCNP has been done to generate data for training of the NN. F1 tally was used along with ACT card in MCNP input file. The ACT card generated gamma lines data resulted by neutron activation of a sample. Samples irradiated in the simulation have a spherical surface geometry in air. F1 tally with relevant tally multiplier can reveal how many particles passing the surface for a given time. More accurate representation of the simulation would be through F8 tally. However, time bins are not allowed with this F8 tally. SSW (Surface Source Write) card also does not have time option to make sequential simulation for the exact experimental procedure. It records not only delayed gamma particles but also prompt gammas which generate unexpected spectra. Since the focus in the simulation is to simulate how many particles are emitted during 8 hours after decay time, F1 tally was chosen for the simulation. In the material card, 260 randomly generated mass concentration cases were prepared.

Simulated peak counts from a gamma spectrum require corrections. It is because experimental data are affected by the detector area, Germanium (Ge) photopeak efficiency, detector efficiency, etc. Although these values can be approximately calculated separately, comparison between experimental and simulated data can reveal an effective total correction. The sample measured in the detector can also be simulated with MCNP. Nevertheless, the simulation assumes perfect conditions. Difference between experimental peak counts and simulated particle counts can be assumed as the correction factor.

3. Neural Network with MATLAB

One of the advanced tools to conduct NN with many features is the MATLAB toolbox. 260 spectra were generated to train the NN in MATLAB. Since number of neurons in the hidden layer affect the system, optimum number can be compared with resulting error. In addition, comparison of LM, BR, and SCG can help to choose best training algorithm for the application. Despite, relative error is being primary criteria, time consumption should also be considered for a large and complicated systems. The determined correction value between simulated data and experimental data has to be applied to the results as NN would not be sensitive to a constant multiplication in the spectrum values.

III. RESULTS

259 spectra generated by MCNP were used to train the network. By default, 70% of data for training, 15% of data for testing, and another 15% of data for validation randomly were divided. One spectrum which contains data from Au, Br, K, Zn, and Na were kept for external validation.

Fig. 2 shows error behavior with number of neurons in the hidden layer in LM backpropagation. Even though, error would be different for every training cases due to randomization, smallest error was found when 23 neurons in the hidden layer were used.

Comparing error dependence in Fig. 2, 3, and 4; LM algorithm served best results for the system. Fig. 3 and 4 reveal higher level of error. Error was different for every training due to randomization on training, testing, and validation data. Behavior was observed to be similar. It was also clear that high number of neurons did not improve the

results. Further optimization was, then, carried out by using LM algorithm in the training part of the NN.

Fig. 5 represents how much input data is required for a certain error level. If error less than 10% was desired, it was found more than 200 prepared spectra are needed for this research. Analysis of more elements in the gamma spectra would require a different number of spectra set.

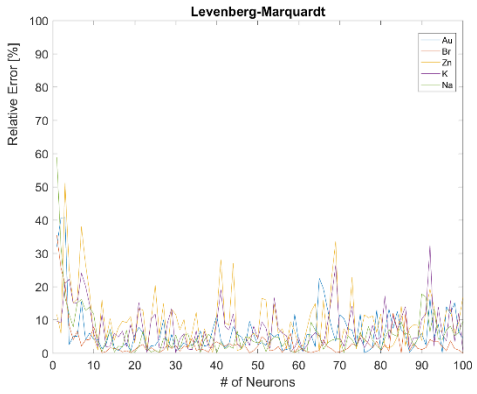

Fig. 2. LM algorithm number of neuron dependence.

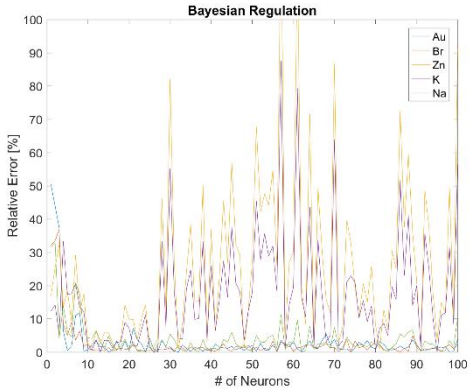

Fig. 3. BR algorithm number of neuron dependence.

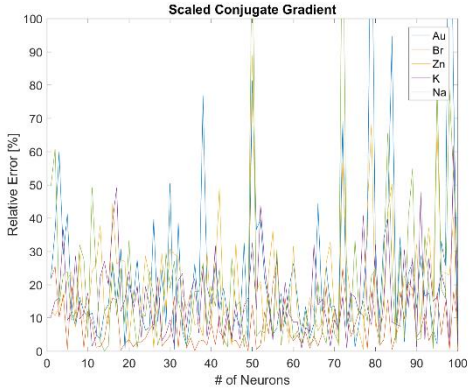

Fig. 4. SCG algorithm number of neuron dependence.

When analysis of a single element spectrum was conducted, the errors for the other four elements were higher than multiple element spectra. A single element analysis resulted negative mass values for non-existing elements in the spectrum. This showed more single element simulation was required to improve the analysis. On the other hand, the kidney stones contained all the elements which were simulated.

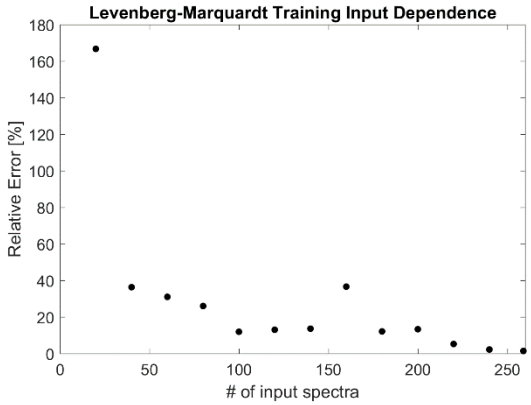

Fig. 5. Error distribution relation to the size of training data with LM algorithm.

Table 1 provides error between actual mass values and mass value calculated with NN. The highest error was observed in Zn concentration. This was attributed to decay half-life of Zn was relatively high compared with the other four.

Table I. Neural Network Error in %

	APA	COM	UA	STRU
Br	5.3	10.1	6.0	14.6
Na	7.2	12.3	8.7	9.0
Zn	17.9	28.2	24.9	27.9
K	-	-	21.1	-
Au	5.2	9.9	5.4	16.1

IV. CONCLUSIONS

MCNP simulation of the irradiation experiment were conducted to generate simulated data. The data were used to train the NN to analyze experimental data. Neural fitting with LM training algorithm was used to optimize the system and analyze experimental spectra.

In order to validate NN, optimization with different training algorithm and number of neurons in hidden layer as well as size of input data were compared. LM algorithm with showed smallest maximum error among five elements. Number of neurons in hidden layer represented better error estimation were observed between 10 to 27. 23 neurons in this research were used. Moreover, for error less than 10%, the size of input should be more than 200 spectra.

The resulted error was less than what has been reported. Thus, it was shown that NN application on NAA can be made successfully. However, there is a limitation caused by the nuclear data library. Training spectrum can be generated by MCNP only if data library is available for certain isotopes.

Highest error was resulted from Zn. This is attributed to its relatively long decay half-life. It can be improved by increasing training data with more cases of Zn.

Although the study showed good errors approaching to desired values, correction factor was dominant source of error. Nevertheless, the cure can be made by simulating actual detector response. In this case, F8 tally used in MCNP is desired when a method for time binning on SSW card can be found. Using a more accurate spectrum as an input would eliminate error propagated from efficiency, photopeak ratio, effective detector area, etc.

Furthermore, this approach setup can be generalized for different experimental setup with no cost except a proper accounting for decay and counting time.

NOMENCLATURE

NAA = Neutron Activation Analysis
NN = Neural Network
MCNP = Monte Carlo N-Particle
HPGe = High Purith Germanium
LM = Levenberg-Marquardt
BR = Bayesian Regularization
SCG = Scaled Conjugate Gradient

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