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A quantitative PGNAA study for use in aqueous solution measurements using Am–Be neutron source and BGO scintillation detector

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ABSTRACT

A prompt gamma neutron activation analysis (PGNAA) system including an Am–Be neutron source and BGO scintillation detector are used for quantitative analysis of bulk samples. Both Monte Carlo-simulated and experimental data are considered as input data libraries for two different procedures based on neural network and least squares methods. The results confirm the feasibility and precision of the proposed methods.

1. Introduction

Neutron activation (NA) is an efficient and non-destructive method for qualitative and quantitative analyses [1]. The method is simply categorized into prompt-gamma neutron activation analysis (PGNAA) where the de-excitation gamma-rays are measured simultaneously with thermal neutron irradiation and delayed-gamma neutron activation analysis (DGNAA) in which the gamma-ray measurement is undertaken after a specific delay time. The PGNAA method is basically used in the analysis of bulk materials such as those performed in cement industries [2], copper [3] and ore [4] mines, salinity measurements [5] and explosive detection [6] with the capability of isotopic analysis. Alternative ways, such as chemical-based and X-ray fluorescence (XRF) methods, are unable to precisely analyze large bulk samples.

This study aims to compare the methods which determine the unknown weight percent of an element in a neutron-activated sample (i.e., quantitative study) using prepared spectrum libraries (generated with the Monte Carlo MCNPX code [7]) and by incorporating a well-known least-squares method and a artificial neural network (ANN) technique [8]. Section 2 of this paper shows that the MCNPX code can be used as a reliable simulation tool for generating the responses of scintillation detectors when exposed to neutron-activated samples. Also a comparison between two commonly-used scintillators, NaI(Tl) and bismuth germinate (BGO), is undertaken in Section 3 to find out which one is more appropriate for the present PGNAA studies. The descriptions for least squares and neural network methods are given in Section 4.

2. PGNAA setup and detector spectra

The measurement setup includes an Am–Be neutron source of around 20 Ci americium activity (equivalent to 4.4E7 neutrons per seconds) that is located inside a 50 cm cylindrical hole in the concrete floor, kept at 30 cm below the sample. A 2 mm lead layer, just on the floor surface, has been used for the reduction of Am–Be 4.43 MeV gamma rays (Fig. 1). In order to prevent the thermal neutrons to enter the scintillation detectors which normally produce large number of ambient gamma rays, two sheets of 5 wt% ^{10}B borated polyethylene (BPE) with $5 \times 50 \times 50$ cm^3 dimensions are used. A relatively thick layer of lead with $5 \times 30 \times 32$ cm^3 dimensions (i.e., thickness, length and width are 5 cm, 32 cm and 30 cm, respectively) is used between the detectors and BPE sheets to shield the detector from background gamma rays. Two gamma-ray detectors, a BGO and an NaI(Tl) scintillators, of right circular cylinder shape and 3″ by 3″ size (i.e., 7.62 cm diameter by 7.62 cm length) are used at both sides of the sample. The whole detection system is surrounded with a wall of 5 cm-thick lead bricks. The prompt gamma-rays from an $NaCl \cdot H_2O$ sample in a cylindrical container is measured with the scintillators in 1800 s live time. The measurement is repeated for 0–500 g NaCl in 3500 cc distilled water (i.e., 51 measurements, including the sample with 0 g NaCl,

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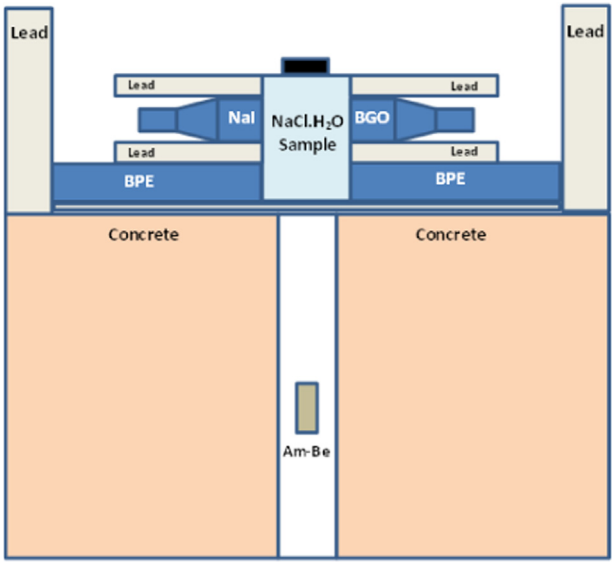

Fig. 1. The schematics of detection setup for use in PGNAA measurements (Dimensions are not scaled).

are performed). Both BGO and NaI scintillator spectra for different NaCl contents are illustrated in Figs. 2 and 3.

Fig. 4 shows a comparison between the responses of BGO and NaI scintillators when exposed to a 3.5 l $NaCl \cdot H_2O$ solution with 500 g NaCl. As seen, the gamma ray peaks are well-resolved in BGO compared to NaI scintillators. The areas under chlorine full-energy peaks of 5.751 MeV and 6.110 MeV may be regarded as a good measure of detection efficiency as illustrated in Fig. 6. Both Figs. 5 and 6 confirm that BGO scintillator represents better performance in PGNAA analyses.

All data analyses and curve fittings have been performed with Origin 9.1 software [9]. For each energy, the counts corresponding to the area within 3σ around the central channel has been taken as peak area. The background subtraction also performed for each energy prior to the peak area calculation. A special feature of Origin software has been used for each peak to subtract the counts below the baseline. This procedure has been repeated for every measurement.

The variation of full-energy peak area for 5.751 MeV and 6.110 MeV gamma rays shown in Fig. 5 may be modeled as following:

$$\text{Area} = a + be^{-\frac{\text{Nacl content}}{c}} \quad (1)$$

where a, b and c parameters have been calculated using Origin as listed in Table 1.

According to (1), the full-energy peak area increases non-linearly with increasing NaCl content, which may be attributed to: (a) the non-linear response of scintillation detectors, (b) the increase in NaCl content when the total solution volume is constant means that HO_2 elemental contents decrease, which results in a somehow unpredictable behavior of the areas under chlorine full-energy peaks. One should know that for low chlorine content, the majority of neutron captures are taken place by hydrogen nuclei as well as a large number of neutron escapes from the solution due to longer neutron mean free path. Whilst, at high chlorine contents, the ^{35}Cl neutron captures are dominant.

Fig. 6 shows the area under the experimental spectra measured with BGO scintillator for an $NaCl \cdot H_2O$ solution of 0 g, 50 g and 100 g NaCl contents. The experimental data and their error bars confirm that the BGO scintillator is capable to well distinguish the samples with 50 g NaCl content difference. The same conclusion may be taken from Fig. 7 for NaI(Tl) scintillator.

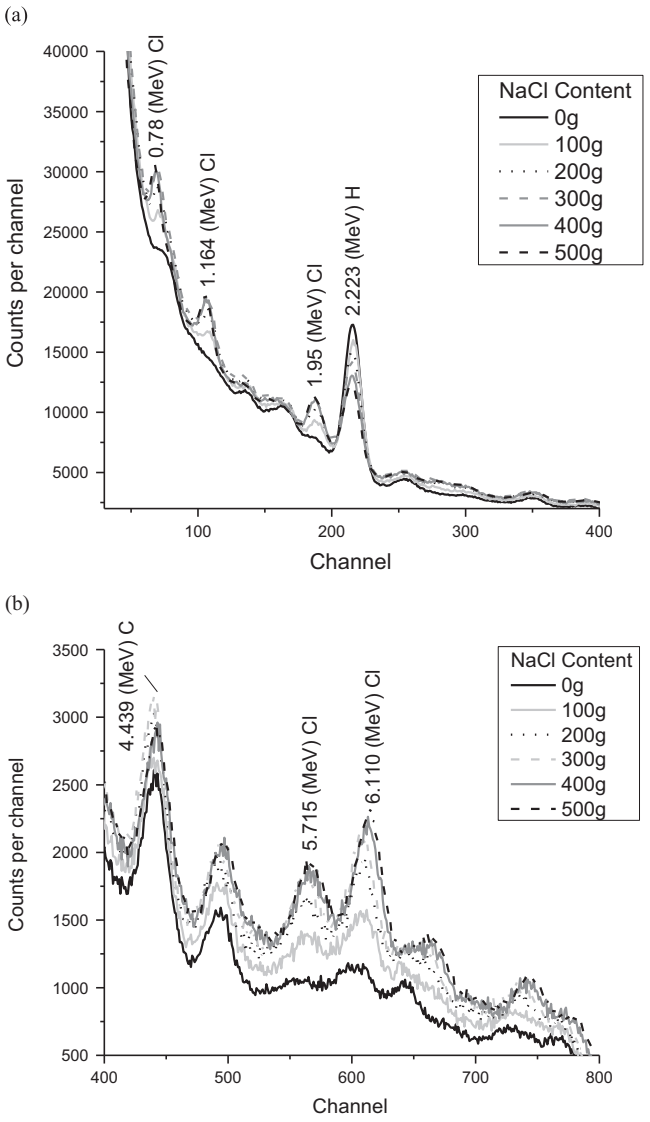

Fig. 2. The BGO scintillator response when exposed to $NaCl \cdot H_2O$ solution of different NaCl contents for gamma-ray energy region of (a) 0–4 MeV. (b) 4–8 MeV (The Channel/MeV ratio has been set to 100).

3. Simulation studies

The measurement setup of Fig. 1 has been simulated with the MCNPX code in neutron–photon mode. The MCNP pulse-height equivalent or F8 tally for BGO and NaI(Tl) scintillators when irradiated with the prompt gamma rays emitted from an $NaCl \cdot H_2O$ solution exposed to an Am–Be source in energy range of 0–11 MeV in 1024 energy intervals are plotted in Figs. 8 and 9. A Gaussian energy broadening, GEB, has been used in the MCNPX input file similar to the work of Miri-Hakimabad et al. [10] to incorporate the contributions of optical photon transport, photomultiplier tube, etc. into the pulse-height resolution for which the broadening function parameters (a, b and c) have been obtained by measurement.

The relatively small difference between experimental and simulation data may be attributed to scattered 4.43 MeV gamma-rays of Am–Be source, the background gamma rays and electronic noise. The insufficient knowledge of material contents in measurement setup (e.g., floor concrete and polyethylene) can be another source of discrepancy. It has been decided to use BGO

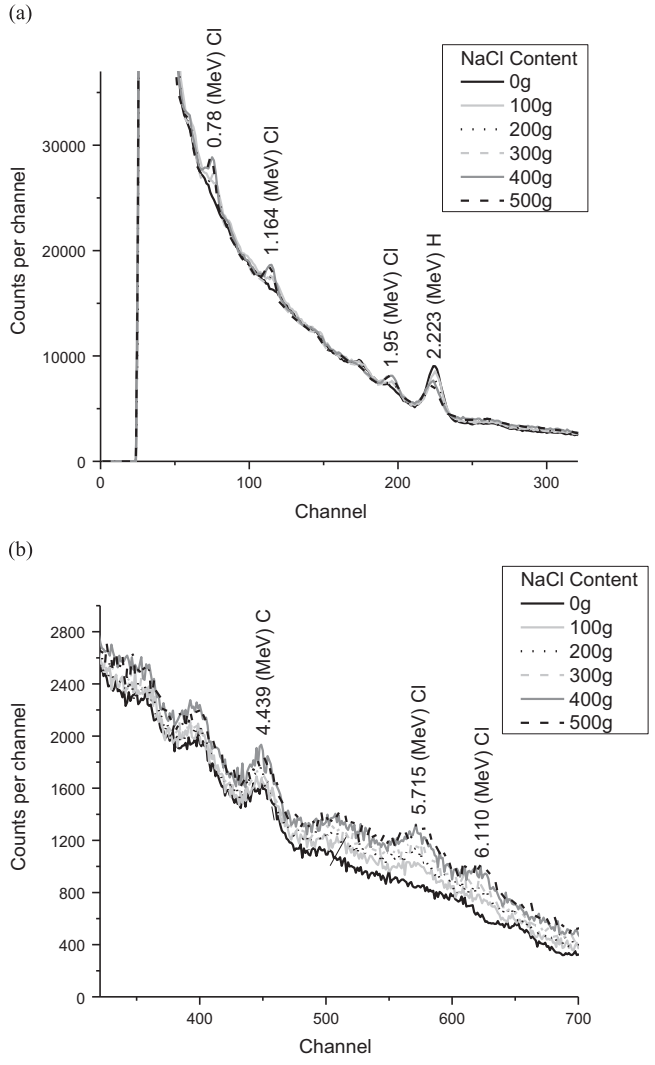

Fig. 3. The NaI(Tl) scintillator response when exposed to $NaCl \cdot H_2O$ solution of different NaCl contents for gamma-ray energy region of (a) 0–3 MeV (b) 3–7 MeV (The Channel/MeV ratio has been set to 100).

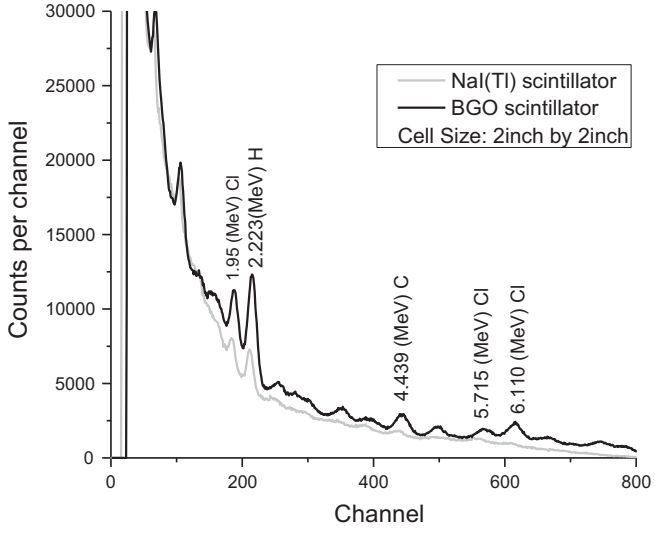

Fig. 4. A comparison between the BGO and NaI(Tl) scintillators responses to a 3.5 l $NaCl \cdot H_2O$ solution (500 g NaCl) (The Channel/MeV ratio has been set to 100).

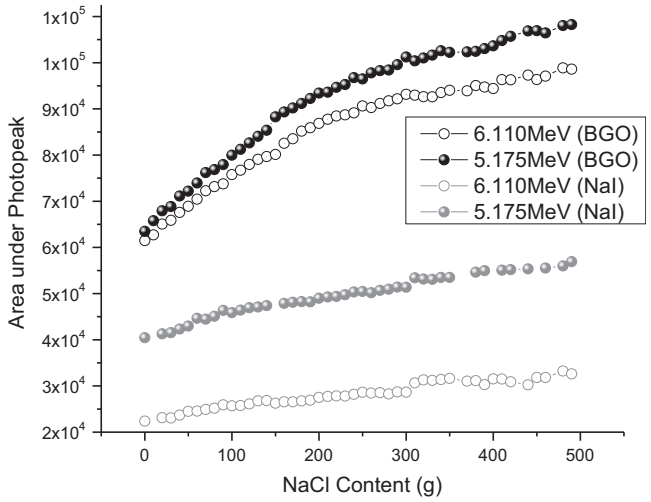

Fig. 5. The areas under the main chlorine full-energy peaks (at 5.715 and 6.110 MeV) for $NaCl \cdot H_2O$ solution measured with BGO and NaI(Tl) scintillators.

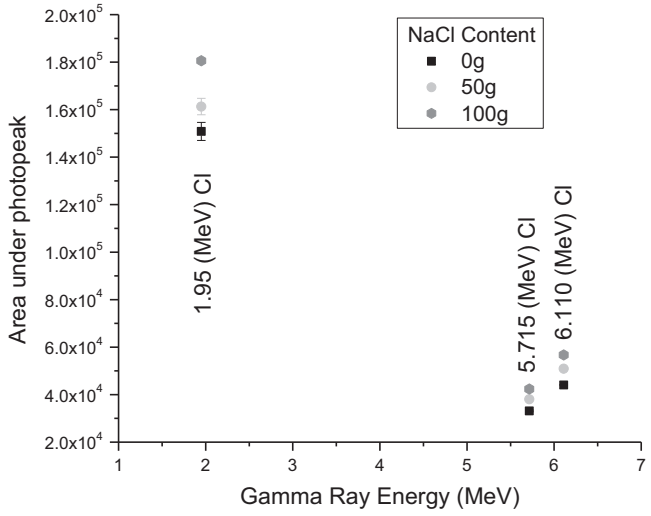

Fig. 6. The areas under chlorine peaks of $NaCl \cdot H_2O$ solution with 0 g, 50 g and 100 g NaCl content measured with BGO scintillator.

scintillator for the rest of studies due to its higher detection efficiency compared to NaI(Tl).

4. PGNAA quantitative analysis

4.1. Neural network method

Having assumed that the MCNPX-simulated spectra give a correct behavior of the PGNAA setup, numerous PGNAA spectra (in 1024 energy bins) have been simulated corresponding to different NaCl contents in $NaCl \cdot H_2O$ solution which are further used in training phase in NAA calculations. At the output layer, the NaCl content of every input spectrum has been inserted in grams (102 spectra corresponding to 102 different NaCl contents), which trains the network how to correlate between the NaCl contents and the peak intensities. The network adjusts the neuron weights using the method of error backpropagation [11], for which 14 different gamma ray spectra, other than those of training phase, have been given for controlling the network. Then, using neural network feature of MATLAB 8.3 Toolbox [12], a multi-layer network (see Fig. 10) including an input layer, a hidden layer and output layer have been constructed. The MATLAB

Table 1
Fitting parameters for the variation of peak area against NaCl content for BGO and NaI scintillators (Fig. 5).

	Photopeak energy (MeV)	Fitting model: $y=a+be^{-\frac{x}{c}}$ Fitting parameters		
		a	*b*	*c*
NaI(Tl)	5.175	41053.05	−18294.27	643.09
	6.110	65309.23	−24407.08	484.06
BGO	5.175	102239.93	−41735.61	212.03
	6.110	113636.00	−50822.48	228.79

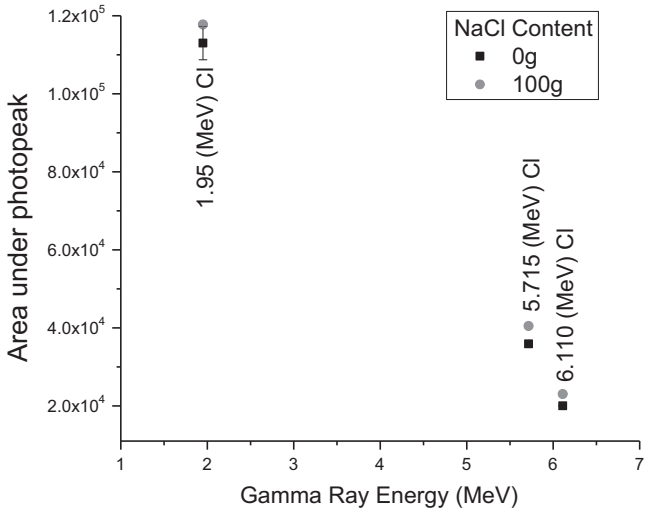

Fig. 7. The areas under chlorine peaks of $NaCl \cdot H_2O$ solution with 0 g and 100 g NaCl content measured with NaI(Tl) scintillator.

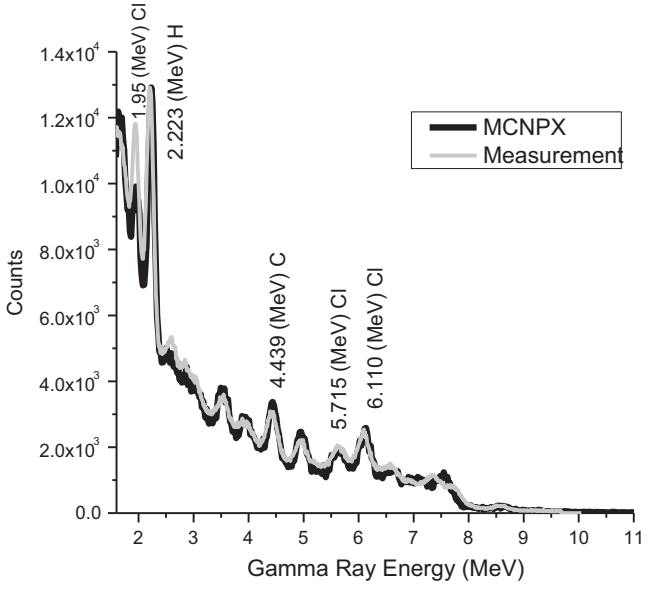

Fig. 8. Experimental vs. MCNPX-simulation data for gamma-ray spectra measured with a BGO scintillator exposed to $H_2O \cdot NaCl$ solution of 500 g NaCl content.

training function `traingdx` for training the network, the transform function `tansig` in the hidden layer and the linear transform function `purelin` for output layer have been incorporated. The input data include 1024 neurons corresponding to the number of channels to specify each spectrum, the hidden layer contains 15 neurons and the output layer contains one neuron. The desired values versus the

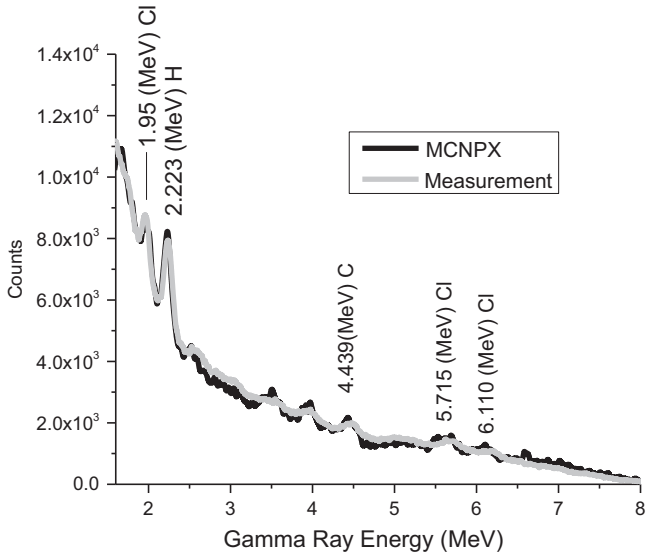

Fig. 9. MCNPX vs. measurement spectra of NaI(Tl) scintillator exposed to $NaCl \cdot H_2O$ sample with 500 g NaCl.

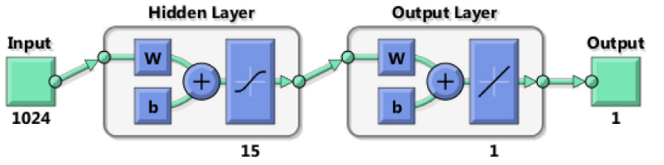

Fig. 10. A neural network design for estimating unknown NaCl content in $NaCl \cdot H_2O$ solution.

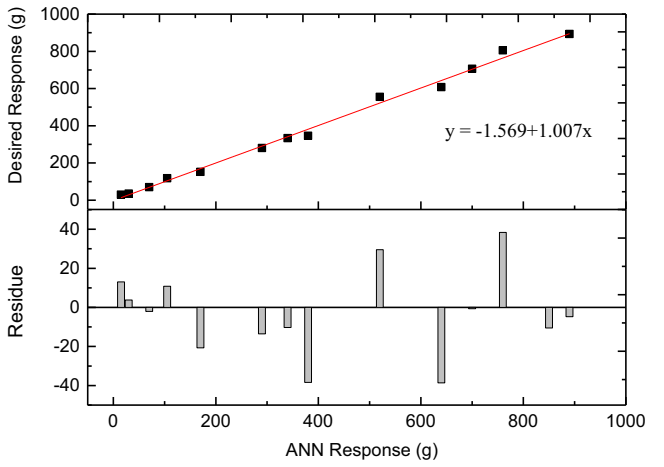

Fig. 11. The correlation between the average values obtained from 14 different measurements and the neural network optimal values. The differences between the data and linear fit have been given as a residue plot.

optimal values of ANN have been illustrated in Fig. 11. As seen, a linear correlation exists between the optimal values and neutron network mean values for the NaCl content in $NaCl \cdot H_2O$ solution.

4.2. Least squares method

In this method, using Monte Carlo MCNPX code, a number of detector spectra corresponding to different NaCl contents in the sample are generated which form the so-called spectrum library. Whenever an unknown spectrum is given, using Eq. (2), the *Y* values are calculated which are the squares of the subtraction values between the unknown, $X_k(i)$, and library spectra, $X_k(j)$,

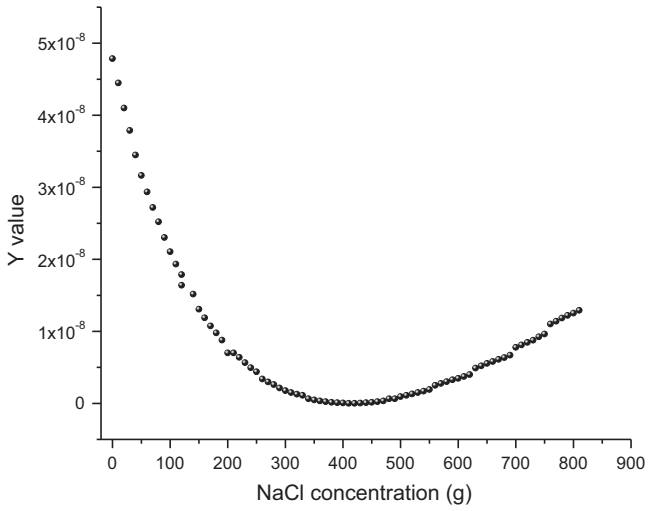

Fig. 12. The summation of squares or Y values against NaCl content in $NaCl \cdot H_2O$ solution where the unknown content is 415 g.

summed over 1024 channels.

$$Y = \sum_{k=1}^{1024} (X_k(i) - X_k(j))^2 \quad (2)$$

The library spectra (i.e., $X(1)$ and $X(2)$) corresponding to two lowest Y values are identified. One may conclude that the unknown spectrum (X) is related to the known spectra following the linear superposition of Eq. (3):

$$X = PX(1) + QX(2) \quad (3)$$

where coefficients P and Q determine the contributions of the two most similar spectra to the unknown spectrum ranging from 0.0 to 1.0.

In this method, using MCNPX code, 82 different library spectra corresponding to different NaCl content (from 0 g to 810 g) in 3.5 l $NaCl \cdot H_2O$ solution have been prepared. The simulations are based on the geometry of Fig. 1 and the spectra are taken with BGO scintillator. A FORTRAN program [13] has been written to take the spectrum of unknown NaCl content as input data and to compare the unknown spectrum with library spectra, channel by channel, to form the summation of squares as in Eq. (2). Clearly, the unknown NaCl content can be found by plotting Y values against NaCl content as explained below.

To investigate the feasibility of the proposed method, the spectrum corresponding to 415 g NaCl in $NaCl \cdot H_2O$ solution has been considered as unknown input spectrum. Fig. 12 illustrates the Y values against NaCl content with a minimum somewhere between 410 g and 420 g. It is assumed that sample contains P value of 410 g and Q value of 420 g content as in Eq. (3). The FORTRAN program gives the values 0.575 and 0.425 for P and Q, respectively. The unknown content is then obtained as 414.25 g with 0.185% relative error.

5. Conclusions

In this research, using experimental and MCNPX-simulated spectra of BGO scintillator when exposed to Am–Be source neutron-irradiated sample have been used to investigate a PGNAA analysis approach for finding unknown content of NaCl in $NaCl \cdot H_2O$ solution. Two different methods based on neural network and least squares have been utilized to find the unknown content which confirms that, in this application, the least squares method has less relative error compared to neural network method.

In most PGNAA applications, the precise gamma-ray peak area determinations are difficult due to adjacent peaks overlap when using relatively poor-resolution detectors such as NaI(Tl) and BGO scintillators. To resolve this problem, the whole prompt gamma ray spectrum (i.e., both photopeak and Compton continuum) has been taken as input data to both least-squares and neural networks analysis programs, in which the information of different peaks of a specific element is regarded as a measure for any change of its content, which is the main advantage of the present study.

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