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MCNP5 Element Library Least Squares Method for On-line Coal Analysis

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ABSTRACT

One of the current methods for the on-line analysis of the coal is the neutron inelastic scattering and thermal-capture analysis (NITA) for non-destructive in situ measurements of the elemental concentration of a bulk sample. A prototype apparatus (including detector, fast neutron source, bulk sample and shielding materials) was simulated by using MCNP5 (Monte Carlo N-Particle) software package. In the material card of MCNP5, the concentration of these elements of the synthetic coal sample was used by taking into consideration of the rates of the lignite coal reserve in Aegean Region of Turkey. The histograms of the relation between number of particles and energy of the gamma were converted to Gaussian distribution by using response functions of the BGO detector. With the separate simulation of MgO, Fe_2O_3, CaO, Al_2O_3, SiO_2, S and C, the element library spectrum was obtained in order to fit the spectrum of synthetic coal sample by using MLR (Multiple Linear Regression) method. By using the relation between the coal sample spectrum and the fitted coal spectrum by MLR, the element library least squares (LLs) method was applied to calculate the elemental contents for the relative coal sample.

Keywords: NITA technique, MCNP5, BGO detector, MLR method.

1. Introduction

Nuclear techniques are widely used for non-destructive in situ measurements of elemental concentration of bulk sample in the mining and energy industries [1]. One of the current analysis methods is the NITA technique which relies on the detection of the gamma rays signatures coming from the neutron inelastic-scattering (NIS) and thermal-capture (TNC) reactions with target material. Usage of the NITA technique makes the detection of the fly ash and Sulfur content possible from TNC, while gamma rays of C and O are produced by NIS [2]. An on-line coal analyzer apparatus included NITA technique can be simulated by using a Monte Carlo computational simulation packages, such as GEANT4 [3,4], MCNP5 [5] and FLUKA [6]. In this paper, the on-line coal analyzer apparatus (including detector fast neutron source, bulk sample and shielding materials) was simulated by using MCNP5 (Monte Carlo N-Particle) software package for the simulation. [7]. Five synthetic coal samples, representing the lignite coal reserve in Aegean Region of Turkey, were prepared covering the range of concentration of Carbon and ash content. These samples were simulated for the prototype geometry and then the histograms obtained from the simulations were then converted to Gaussian distribution by using BGO detector response functions [8]. In order to calibrate the spectrum of the synthetic coal sample, MgO, Fe_2O_3, CaO, Al_2O_3, SiO_2, S and C were simulated later in the spherical fixed geometry and the spectrum of each element was used to generate the element library spectra.

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2. MCNP5 simulation of the coal sample

MCNP5 is a powerful general-purpose modeling tool providing continuous-energy, generalized geometry, time-dependent Monte Carlo transport code. It can handle such cases as neutron only, photon only, electron only, combined neutron/photon or photon/electron transport according to characteristics of the problem of interest [9]. The process aimed to simulate the geometry of the prototype apparatus and interaction of the synthetic coal material with fast and slow neutrons, emitting 10^9 neutrons as a total number of particles by using MCNP5. In the simulation, a 14 MeV point source of neutrons which represented a D-T neutron generator of prototype apparatus were generated isotropically just above the sample container, indicated by a red dot in the schematic view of the virtual experimental set up shown in Fig. 1.

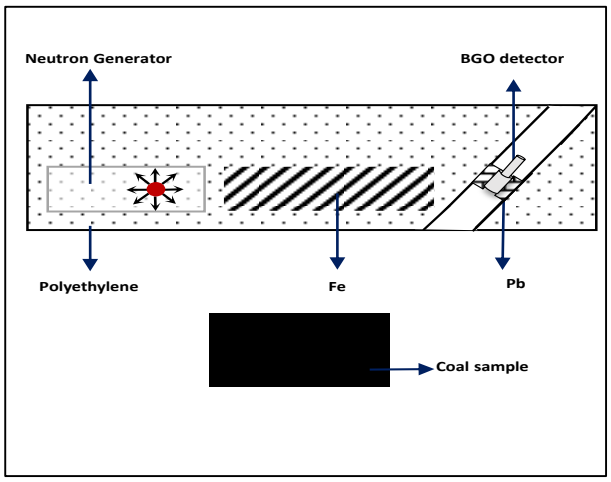

Fig.1 *The schematic view of the MCNP5 simulation geometry.*

The MCNP5 average cell flux tally (or "F4 tally") was used to detect the energies of gamma rays between 0 and 10 MeV with 1024 channels in BGO volume. The bulk material as the combination of MgO, Fe_2O_3, CaO, Al_2O_3, SiO_2, S and C weighted 110 kg in the coal sample volume. The concentration of compounds in each set is shown in Table 1. The five synthetic coal samples including different concentration of compounds were simulated by using MCNP5 simulation package.

Table 1. *The concentration of the compounds in each coal set.*

Compound	Set 1 (%)	Set 2 (%)	Set 3 (%)	Set 4 (%)	Set 5 (%)
C	76.00	69.34	62.66	56.00	49.34
SiO_2	8.20	10.86	13.60	16.34	19.00
Fe_2O_3	3.60	4.6	5.86	7.10	8.22
Al_2O_3	3.60	4.94	6.14	7.34	8.62
MgO	1.00	1.26	1.60	1.86	2.20
CaO	3.60	4.94	6.14	7.34	8.62
S	4.00	4.00	4.00	4.00	4.00

Additionally, the MCNP simulation of background of prototype apparatus was also performed by using F4 tally with 10^9 neutrons as a total number of particles. The histogram results of the simulations were converted to spectrum by using BGO detector response functions, which are obtained experimentally. For the first coal sample (Set-1), the gamma rays spectrum of MCNP5 simulations is shown in Fig. 2. The upper panel shows foreground and background spectra and the lower panel the net spectrum is shown for the Coal Set-1.

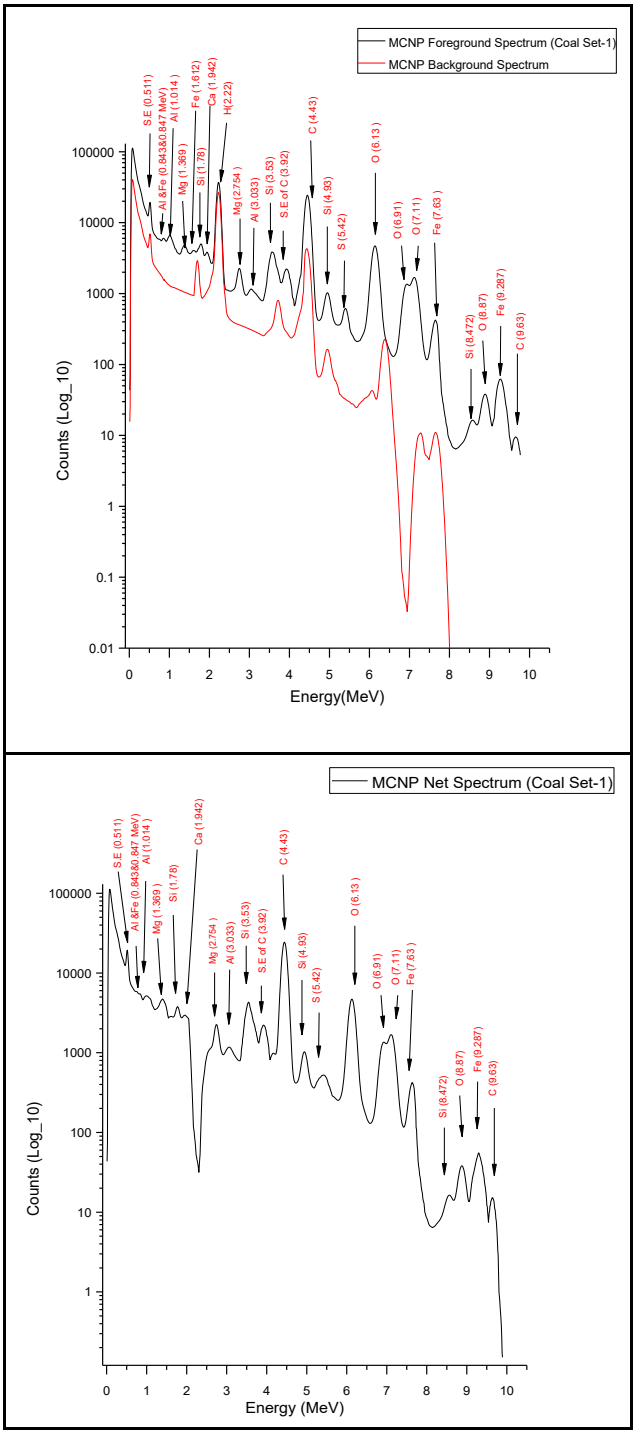

Fig.2 *MCNP5 gamma rays spectrum of Coal Set-1.*

The comparison of five coal sets of the foreground spectrum is presented in Fig. 3.

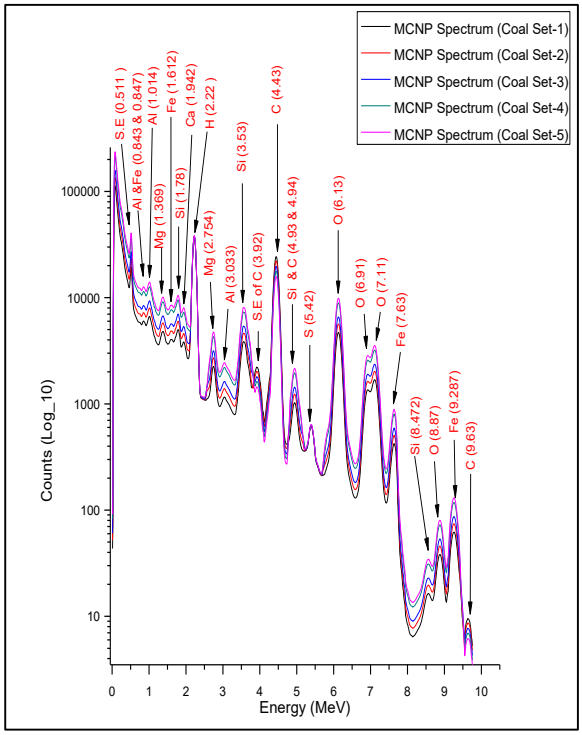

Fig.3 *Comparison of spectrum of 5 coal sets.*

3. Generating of the element library spectrum

Unlike the geometry of prototype apparatus, each of MgO, Fe_2O_3, CaO, Al_2O_3, SiO_2, S, and C compounds was simulated separately in a spherical fixed volume shown in Fig. 4. The point neutron source of 14 MeV was inserted inside the cell#1, which was filled with air medium. The cell#2 was filled with polyethylene material in order to thermalize the neutrons.

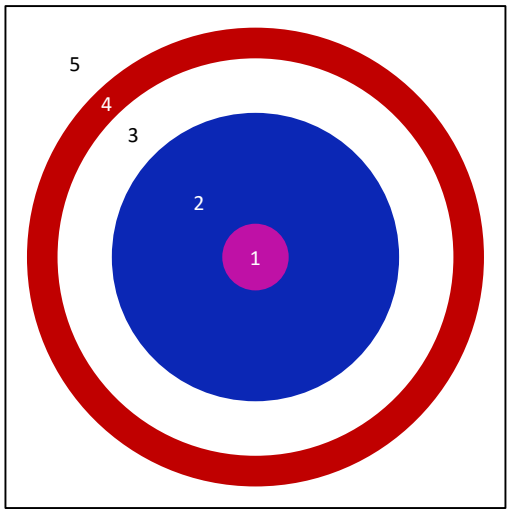

Fig.4 *The spherical fixed-geometry for elemental library spectra.*

The compounds listed in Table-1 were inserted separately to the cell#3 and whole volume was surrounded with BGO detector material inside the cell#4, and the cell#5 represents the vacuum. For this purpose, the MCNP5 average cell flux tally (or "F4 tally") was used to obtain library of the single element spectrum. The particle history of the simulation in nps card was entered 108 as the total number of particles of the neutron source. The histograms and spectrums of the MCNP5 simulation of MgO, Fe_2O_3, CaO, Al_2O_3, SiO_2, S, and C in the fixed spherical geometry are shown in Figs 5 to 11.

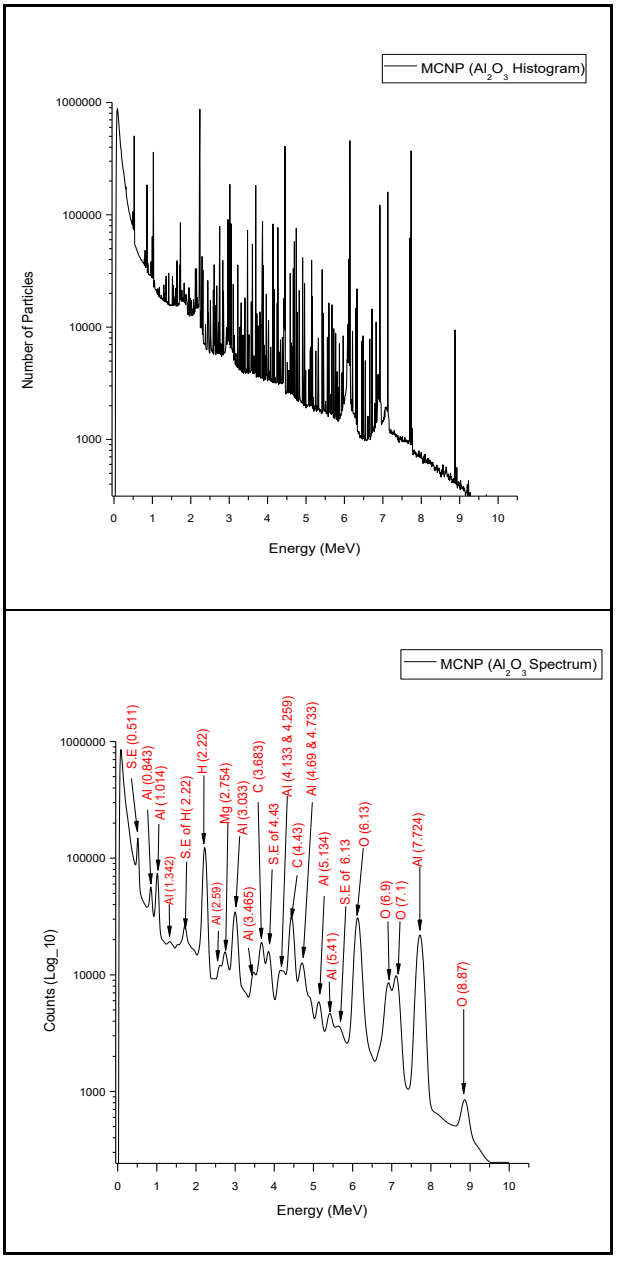

Fig.5 *MCNP5 Histogram and spectrum of Al_2O_3 compound.*

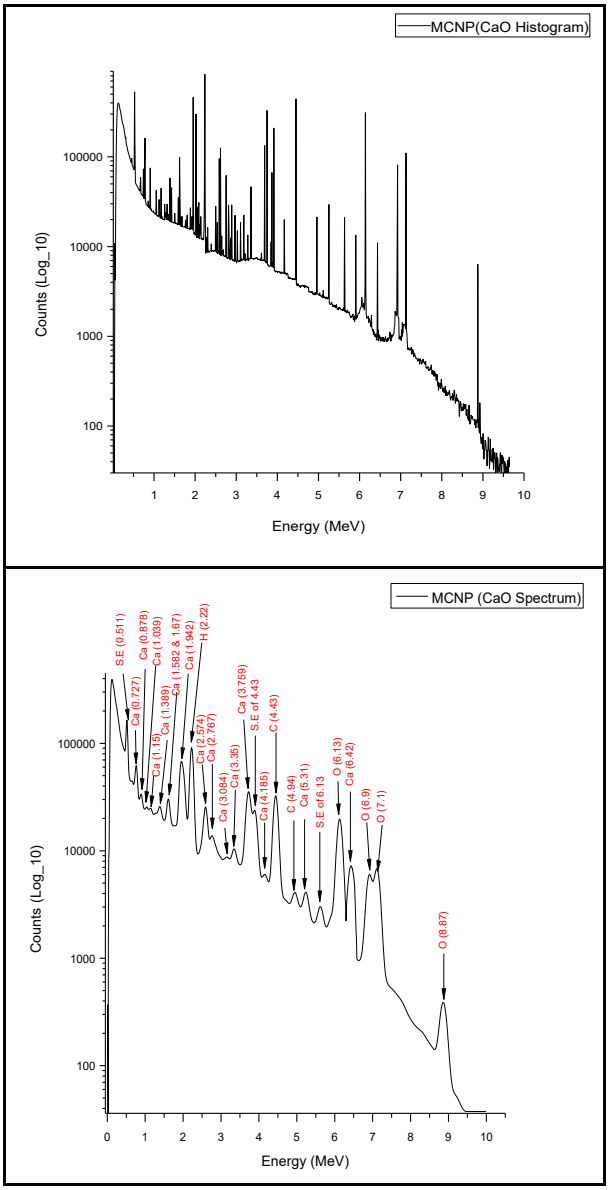

Fig.6 *MCNP5 Histogram and spectrum of CaO compound.*

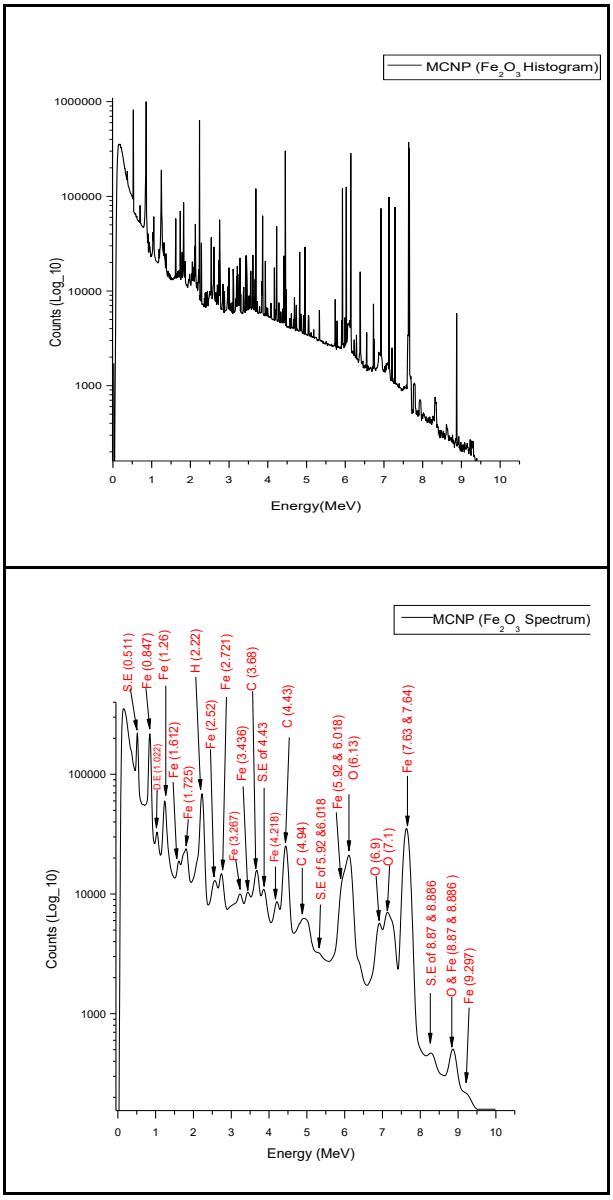

Fig.7 *MCNP5 Histogram and spectrum of* Fe_2O_3.

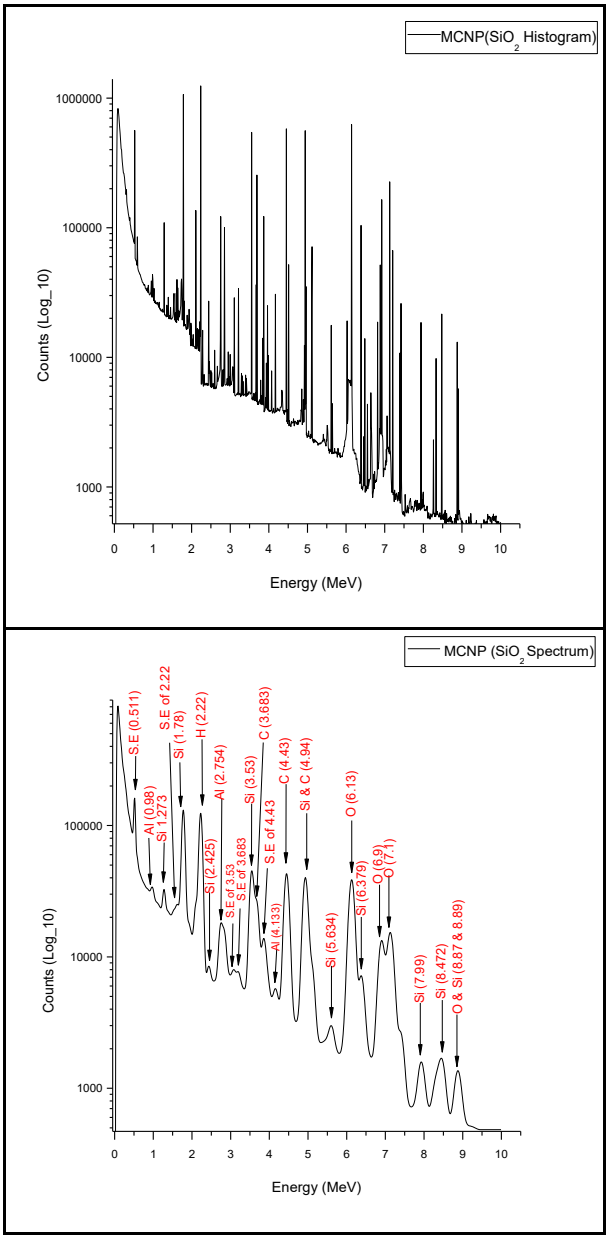

Fig.8 *MCNP5 Histogram and spectrum of SiO_2 compound.*

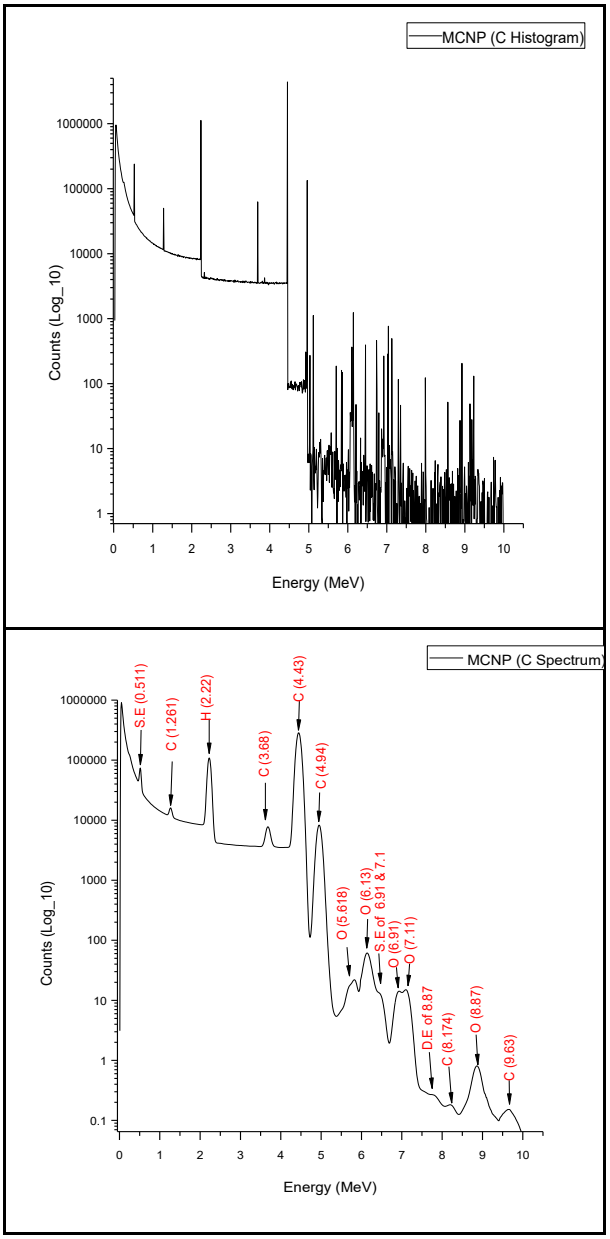

Fig.9 *MCNP5 Histogram and spectrum of the Carbon element.*

Table 2. *The regression variables of compounds and regression coefficients of each coal set in MLR.*

Variable	Set 1	Set 2	Set 3	Set 4	Set 5
a_1 (SiO_2)	31.67304	34.4954	39.61692	54.20798	59.30413
a_2 (Al_2O_3)	38.30900	69.55653	96.52681	161.82605	188.75622
a_3 (CaO)	71.79926	74.14684	76.65868	79.76044	82.58825
a_4 (C)	11.92658	3.28514	-1.18583	-8.81317	-12.83898
a_5 (Fe_2O_3)	-25.27001	-19.3327	-16.53293	-15.21516	-13.21699
a_6 (MgO)	19.23565	27.67508	35.54155	54.98948	62.67590
a_7 (S)	-119.463	-134.307	-159.5405	-2345026	-261.06018
Regression Coefficient	0.990594	0.995270	0.996983	0.997562	0.998977

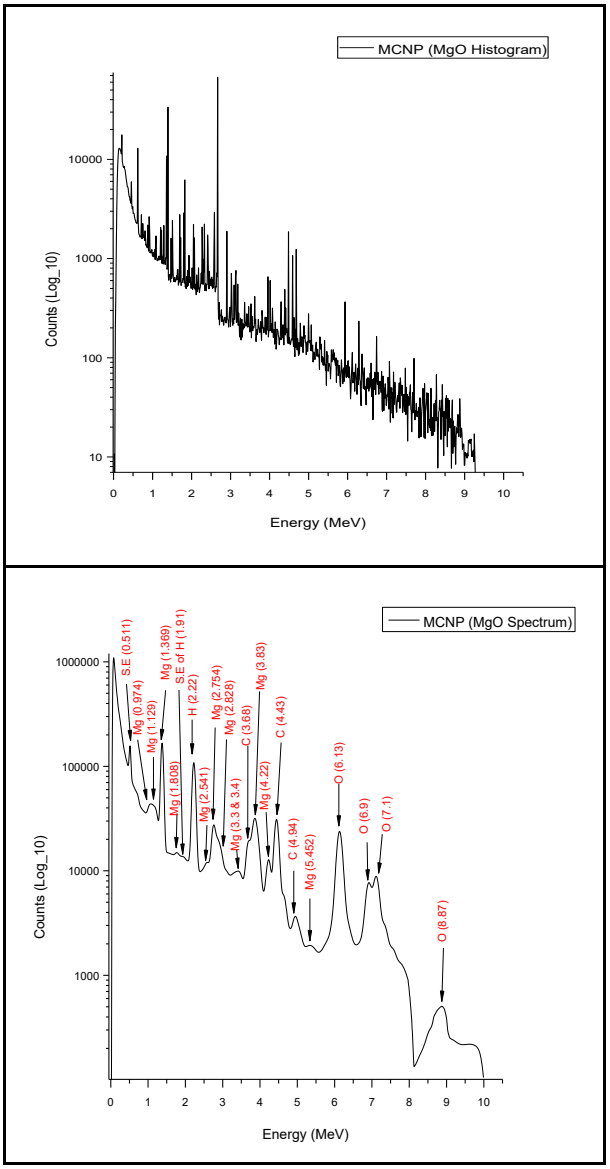

Fig.10 *MCNP5 Histogram and spectrum of MgO compound.*

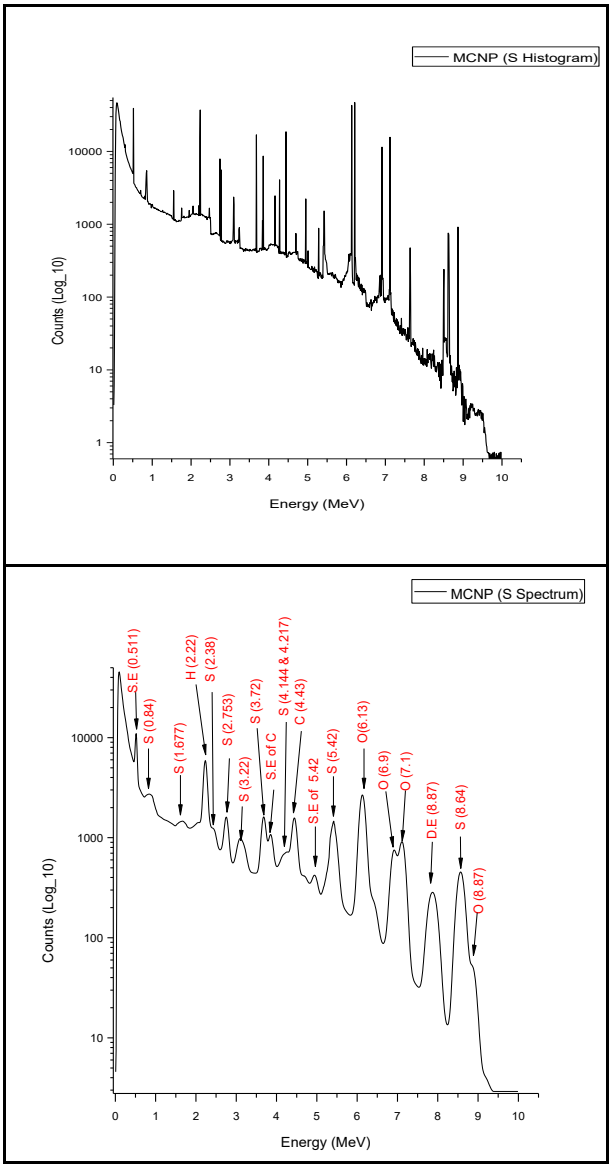

Fig.11 *MCNP5 Histogram and spectrum of S element.*

Table 3. *The comparison of the concentration of elemental contents between prepared and calculated values for each coal set.*

	Net Coal Spectrum Set-1		Net Coal Spectrum Set-2		Net Coal Spectrum Set-3		Net Coal Spectrum Set-4		Net Coal Spectrum Set-5	
Compound	**Prepared (%)**	**Calculated (%)**	**Prepared (%)**	**Calculated (%)**	**Prepared (%)**	**Calculated (%)**	**Prepared (%)**	**Calculated (%)**	**Prepared (%)**	**Calculated (%)**
SiO_2	8.20	8.07	10.86	10.77	13.60	13.75	16.34	16.44	19.00	19.02
Al_2O_3	3.60	3.56	4.94	4.87	6.14	6.22	7.34	7.40	8.62	8.61
CaO	3.60	3.56	4.94	4.87	6.14	6.23	7.34	7.40	8.62	8.61
C	76.0	75.98	69.34	69.26	62.66	62.72	56.0	56.05	49.34	49.34
Fe_2O_3	3.60	3.51	4.66	4.63	5.86	5.91	7.10	7.10	8.22	8.21
MgO	1.00	0.93	1.26	1.25	1.60	1.61	1.86	1.87	2.20	2.20
S	4.00	3.99	4.00	3.98	4.00	4.02	4.00	4.05	4.00	3.98
Total	100.00	99.60	100.00	99.63	100.00	100.46	100.00	100.31	100.00	99.97

Each one of the spectra for the chemical compounds was used to obtain the element library by utilizing Multiple Linear Regression (MLR) method. MLR is a very useful mathematical method to represent any unknown coal spectrum as the sum of relative elemental contents. Mathematical formulation of the MLR was given below in Eq. 1,

$$y_i = yf_i - yb_i = \sum_{j=1}^{n} a_{ij}x_j, \quad i = 1, 2, 3 \ldots 1024 \qquad (1)$$

where the parameter yf_i is the count rate on i^{th} channel for the foreground spectrum of the unknown coal sample, yb_i is the background spectrum belonging to prototype apparatus, a_{ij} is the count rate of the net spectrum at the ith channel for the relative element library in the spherical geometry for j^{th} element, x_j is the relative content of j^{th} element. The net spectrum, y_i was obtained by subtracting the background spectrum yb_i from the foreground spectrum yf_i as shown in Fig.2. After having applied the MLR method on the net coal spectrum y_i, the MLR and Coal Set-1 spectra were shown in Fig.12.

The regression coefficients obtained from MLR and the regression variables of compounds of each coal set were shown in Table 2.

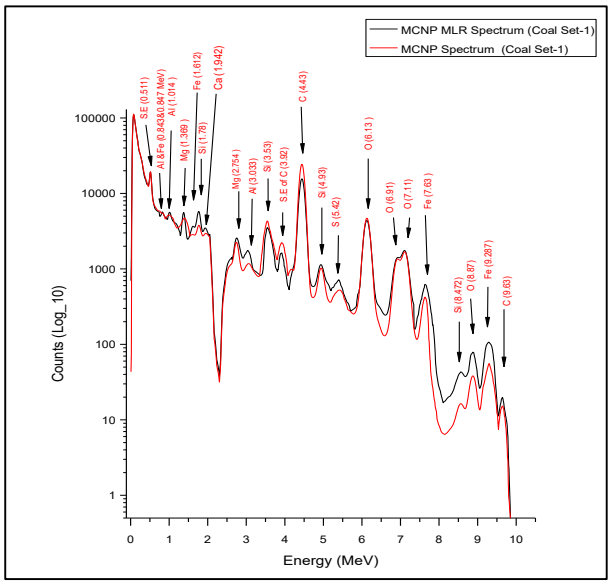

Fig.12 *Comparison the MCNP5 Spectrum of Coal Set-1 with Coal Set-1 spectrum obtained from MLR method.*

Since the desirable correlation between the net spectrum and the combination of element library is reached, the concentrations of the elemental contents for the related coal sample can now be calculated by using library least squares (LLs) method as given by Eq. 2.

$$M = \sum_{i=1}^{1024}\left(y_i - \sum_{k=1}^{n} a_{ik}x_j\right)^2 \qquad (2)$$

where M is the most probable value for elemental contents of the given related net coal spectrum.

By assuming the a_{ij} is constant, the partial derivative of the term of M with respect to the elemental contents shown by x_j is set to zero given by Eq. 3 [10].

$$\frac{\partial M}{\partial x_j} = \sum_{i=1}^{1024}\left(y_i - \sum_{k=1}^{n} a_{ij}x_j\right) a_{ij} = 0 \qquad (3)$$

The matrix form of the Eq. 3 was shown in Eq. 4.

In order to calculate the concentrations of elemental contents, the Eq. 5 was solved for X.

$$A^T Y - (A^T A)X = 0 \qquad (4)$$

$$X = (A^T A)^{-1} A^T Y \qquad (5)$$

where **X** is matrix the elemental contents of the coal spectrum, **A** is the matrix of the combination of the single element library obtained from MCNP simulation, **Y** is the matrix of the related net coal spectrum, $\mathbf{A^T}$ is the transpose matrix of **A**, $(\mathbf{A^T A})^{-1}$is the inverse matrix of the multiplication of $\boldsymbol{A^T}$ and **A**. By using Eq. 5, the **X** matrix was calculated for each coal spectrum, and the results showing good agreement between prepared elemental contents and calculated values are given in Table 3. It should be noted that the deviations are less than 0.5% from the prepared values of the elemental contents.

4. Results and conclusion

The prototype set-up was designed for detecting the content of elements in coal sample by using the NITA technique. The simulation results of coal sample sets were consistent with Carbon, Sulfur and ash contents in Table 1. From coal sample set 1 to set 5 respectively, the photo peak level of neutron induced gamma energy representing the carbon content was decreased, whereas gamma energies of elements of Ca, Fe, Si, Al, Mg in ash content were observed to increase. According to the simulation of the coal spectrum, gammas can be detected easily due to high reaction cross section at these gamma energies; C (4.43 MeV), O (6.13 MeV), Fe (7.63 MeV), Si (3.53 MeV), Ca (1.942 MeV), Al (1.014 MeV), Mg (1.369 MeV), S (5.42 MeV), H (2.22 MeV). Some of the elements such as S (0.84 MeV), Al (0.843 MeV) and Fe (0.847 MeV) which gamma energies are close to each other cannot be detected due to the low resolution of the BGO detector.

The correlation factor between the MLR and the coal sample spectrum was found greater than 0.99 for each coal sample. This result shows that the simulation of coal sample is consistent with the simulation of the elemental library. Since desirably good correlation is reached, this can allow us to calculate the element contents of any coal sample by using library least squares (LLs) method. According to Table 3, after using the LLs method, the calculated value of the concentration of elements was

found in excellent agreement with the prepared value of the concentration of the elements.

We will consider other elements and multi-elemental measurements in the following-up studies. Further investigations must be performed to obtain elemental library of experimental spectra to determine the element contents.

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Conflicts of Interest

The authors have no conflict of interest.

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