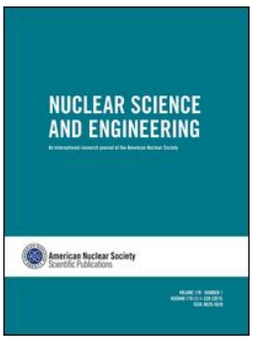

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Computer Code Abstract

CEARPGA II: A Monte Carlo Simulation Code for Prompt-Gamma-Ray Neutron Activation Analysis

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Abstract—*A Monte Carlo simulation code, CEARPGA II, has been developed to generate the complete set of library spectra that are required for the application of the Monte Carlo–Library Least-Squares approach in prompt-gamma-ray neutron activation analysis. Compared to the previous version, the CEARPGA I code, several important improvements have been made including eliminating the "big weight" problem by implementing the Analog Linear Interpolation technique, generating the appropriate detector response functions using improved simulation models that account for NaI detector nonlinearity and flat continua, generating the neutron activation backgrounds by directly sampling detector-activated gamma-ray energies, generating the natural background libraries by interpolating the energy-score tables, and tracking the annihilation gamma rays from the pair production interaction that occurs outside the detector. The coal sample spectrum calculated with the CEARPGA II code is benchmarked against those calculated from the CEARPGA I code, the MCNP code, and experimentally measured data.*

I. INTRODUCTION

The Monte Carlo–Library Least-Squares (MCLLS) algorithm has been proposed for the elemental analysis of sample materials in prompt-gamma-ray neutron activation analysis[1–3] (PGNAA). It is capable of accounting for the basic nonlinearity of the inverse PGNAA approach. For this purpose, the CEARPGA I Monte Carlo code was developed within the Center for Engineering Applications of Radioisotopes to generate the required elemental library spectra. The system originally modeled was a PGNAA coal analyzer, which consisted of a ^{252}Cf neutron source and a Ge detector. The code incorporated a variety of variance reduction techniques to improve its calculation efficiency such as forcing all prompt gamma rays to be emitted after an (n,γ) interaction has occurred (stratified sampling), using the expected value splitting technique to increase the scoring probability of each gamma ray that is tracked, using the correlated sampling method to deal with small variations of compositions in the sample, and using detector response functions to convert the gamma-ray spectra incident on the detector to pulse-height spectra. This code generated not only the elemental library spectra based on any given composition of a sample but also the backgrounds from the neutron source and those from the structural materials of the device.

Although the CEARPGA I code was successfully used to implement the MCLLS algorithm for coal samples, some improvements were required. The primary problem was the "big weight" problem that was observed within the lower range of simulated spectra.[3] Figure 1 shows an experimental coal spectrum and the corresponding CEARPGA I code–simulated spectrum. It is found that there exist several strange peaks in the position near channel 219 (at ~1.214 MeV) of the simulated spectrum that are not observed in the experimental spectrum. This indicates that there exist some big weights in the particle histories that affect the estimate of the mean of these data so that the resulting mean estimate does not converge to the true value, even though the number of simulation histories is sufficient for a good estimate.

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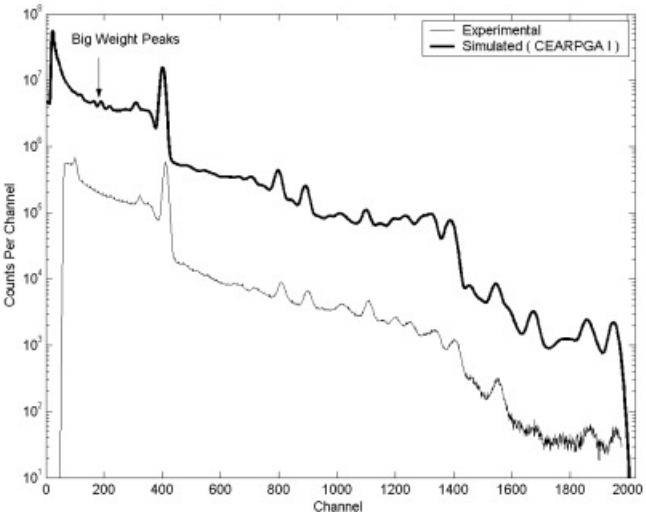

Fig. 1. CEARPGA I–calculated coal spectrum and experimental coal spectrum.

The expected value splitting approach was identified as being responsible for the big weight problem.[3] In this approach, the gamma ray being tracked is split into two parts at any interaction site: One part is forced directly to the detector and detected without further interaction, and the other part with the rest of the weight of the original particle undergoes the normal scattering process. The detected probability of the first split gamma ray can be calculated from the following formula[1]:

$$P = \int_{\upsilon_{\min}}^{\upsilon_{mas}} \int_{\rho_{\min(\upsilon)}}^{\rho_{\max(\upsilon)}} p_1(\upsilon,\rho) \times \rho_2(\upsilon,\rho) \times p_3(\upsilon,\rho)\, d\rho\, d\upsilon \ , \quad (1)$$

where

p_1 = probability of scattering toward the detector through the angles (υ,ρ)

p_2 = probability that the gamma ray will be transmitted to the detector at the direction angles (υ,ρ) without interaction

p_3 = total detection efficiency of the detector from a gamma ray incident on the detector through the (υ,ρ) angles.

It is understandable that those gamma rays generated near the detector have very large angles subtended by the detector, very short flight distances to the detector, and therefore very large resulting probabilities for reaching the detector without interaction and thus can lead to occasional intensities in the total spectra and elemental libraries that are much larger than the average—this is the big weight problem.

The second problem is the response nonlinearity of the NaI detectors,[4–12] which results in the pulse-height energy of the gamma ray not being proportional to the incident gamma-ray energy.

The third problem involves the background spectra.[13,14] The background libraries such as NaI neutron activation libraries and natural background cannot be generated in the CEARPGA I code.

The last problem is that the 0.511-MeV annihilation gamma rays that are generated from pair production interactions outside the detector were not tracked in the CEARPGA I code because the minimum cutoff energy previously used in spectral analysis was >1 MeV. The addition of tracking these gamma rays would make it possible to take full advantage of the information in the entire spectrum.

Therefore, some new approaches have been proposed and are implemented in the CEARPGA II code to solve these problems while increasing both the efficiency and accuracy of the code.

II. MAJOR FEATURES OF THE CEARPGA II CODE

The features inherited from the CEARPGA I code included

1. use of the stratified sampling approach to force all gamma rays to be emitted for the neutron source, the radiative capture interactions, and the neutron inelastic interactions
2. use of the detector response functions to convert incident gamma-ray spectra to pulse-height library spectra to save the time required for tracking the transport of gamma rays inside the detector and to provide better accuracy
3. use of the correlated sampling technique to predict the effect of small variations of sample compositions
4. use of the principle of forcing neutron interactions before they reach the outer boundary to prevent their premature loss.

Additional CEARPGA II code-specific features are described below.

II.A. Generation of NaI Detector Response Functions

A specific-purpose Monte Carlo simulation code, the g03 code, developed specifically for generating detector response functions of NaI detectors,[13] has been incorporated into the CEARPGA II code to generate problem-specific detector response functions. The simulated detector response functions are spread according to a Gaussian distribution with the standard deviation $\sigma_N(E_I)$ given by[13]

$$\sigma_N(E_I) = \sqrt{[\sigma_T^2(E_I) - \sigma_I^2(E_I)]}$$

and

$$\sigma_T(E_I) = aE_I^b \ , \quad (2)$$

where

$\sigma_T(E_I)$ = total standard deviation of the NaI detector response for the gamma ray with incident energy E_I

a, b = fitted model parameters obtained by least-squares analysis of experimental data. For example, for a 7.62- × 7.62-cm NaI detector, a and b were equal to 0.03027 and 0.6593, respectively, while for a 15.24- × 15.24-cm NaI detector, a and b were 0.02609 and 0.6027, respectively, and for a 7.62- × 15.24-cm NaI detector, a and b were 0.0390513 and 0.408146, respectively.

$\sigma_I(E_I)$ = standard deviation associated with the NaI detector inherent nonlinearity, which can be derived by simulating the response of the incident gamma ray for the NaI detector of the infinite size.

II.B. Correction for the NaI Nonlinearity Problem

The nonlinearity of NaI detectors is caused by nonuniform scintillation efficiencies with deposited electron energies inside the detectors and exhibits different values for the full energy peak and the first escape peak.[4–12] This problem was corrected in the calculation of the detector response functions by simply introducing a relative scintillation efficiency factor to multiply each individual deposited electron energy as it occurs within a normal history.[13] The scintillation efficiency factor is calculated from the experimental model[10,11]:

$$S(E_e) = 1 + k_1 \exp[-(\ln E_e - k_2)^2/k_3] \ , \quad \text{for } E_e \geq 10$$

and

$$S(E_e) = 1 + k_1 \exp[-(\ln E_e - k_2)^2/k_4] \ , \quad \text{for } E_e \leq 10 \ , \quad (3)$$

where

$S(E_e)$ = relative NaI scintillation efficiency for a deposited electron energy

E_e = electron energy (keV)

$k_1 = 0.245$

$k_2 = 2.30258$

$k_3 = 7.1635$

$k_4 = 5.1964$.

II.C. Correction for the NaI Detector Variable Flat Continuum Problem

The flat continuum is thought to be caused either by the loss of electrons that escape from the detector surfaces without depositing all of their energy or by a fraction of the electrons that are produced being captured at impurity sites for a period that precludes their contributing to a pulse.[13,15] In practice, the flat continua are detector dependent, and currently, there are no good models to directly predict their intensity. Some simulated flat continuum results predicted by the electron losses from the detector surfaces gave the correct shape according to the normal Monte Carlo simulation models but were as much as an order of magnitude smaller than what was observed experimentally. A possible explanation for this is electron channeling in the single-crystal detectors, which would produce larger losses than normal electron transport. To fix this problem, a semiempirical approach has been devised for use with CEARPGA II that uses a pseudo electron range by a trial-and-error fit to experimental results during the process of calculating detector response functions.[13]

II.D. Use of the Analog Linear Interpolation Approach

The analog linear interpolation approach[16,17] combining both the analog Monte Carlo simulation method and the linear interpolation technique is implemented in the code to eliminate the big weight problem while maintaining the high efficiency and accuracy of the simulation.

II.E. Simulation of Natural Backgrounds

The natural backgrounds include the three libraries from the ^{40}K, uranium, and thorium decay chains, respectively. They are obtained not by tracking their gamma rays step by step but by interpolating the pseudo-gamma-ray energy-score tables established in the simulation. This saves much computational effort.

II.F. Simulation of NaI Detector Neutron Activation

The activation of NaI crystal results in the generation of the prompt gamma rays from both I and Na nuclei and the decay gamma rays from the radioisotopes ^{128}I and ^{24}Na. Three activation library spectra for NaI detectors of different sizes were measured experimentally,[18] including

1. the prompt-gamma-ray spectrum of both I and Na
2. the ^{24}Na decay spectrum
3. the ^{128}I decay spectrum.

They were previously incorporated directly into the CEARPGA I code and used for MCLLS analysis.[17] Monte

Carlo models of these libraries could not be used at that time because of the lack of complete coincidence scheme data.[19]

In the present CEARPGA II code, given the available coincident scheme data, these three libraries can be calculated directly by sampling the deposited gamma-ray energy of any coincident gamma ray from its corresponding detector response function. Compared to the usual Monte Carlo approach, this method has the advantage of accounting for the nonlinearity of the NaI detector because of the use of detector response functions and thus saving much time in tracking each coincident gamma ray.

III. NEUTRON TRANSPORT

III.A. Neutron Cross Sections

The neutron cross-section data used in the code are based on the ENDF/B-V discrete neutron cross-section data file, which covers the neutron energy range from 10^{-5} eV to 20 MeV for all elements at 300 K. All cross sections except the inelastic scattering cross sections are fitted using the cubic spline method on the logarithmic scale over the thermal energy range and the resonance region.[1] The scattering cross sections of hydrogen at neutron energies <2 eV are directly calculated from the following relationships fitted by Yuan, Gardner, and Verghese[20] based on the experimental data for water at 296 K (Ref. 21):

$$\sigma_s = \begin{cases} 21.76255 - 2.84374\zeta + 9.85645\zeta^2 \\ \qquad\qquad 0.0363 < E \le 2.0 \\ -7.86801 - 37.95304\zeta \\ \qquad\qquad 0.0062 < E \le 0.0363 \\ -34.77877 - 49.61637\zeta \\ \qquad\qquad E \le 0.0062, \end{cases} \tag{4}$$

where $\zeta = \ln E$ (E in the unit of electron volts)

III.B. Tracking Neutron Movements

In the CEARPGA II code, each neutron history starts from the neutron source (default ^{252}Cf) where initial neutron energy and direction are sampled. The neutrons emitted directly from the neutron source are assumed to be isotropic. The flight path length is sampled based on a normal exponential function.

Among all types of the neutron interactions, the inelastic scattering interaction, elastic scattering interaction, and radiative capture interaction are sampled explicitly while the other interaction types such as the (n,α) reaction, etc., are treated implicitly by adjusting neutron accumulated weights accordingly. Three different techniques are adopted in sampling the interaction type depending on the specific situation. The first one is applied to sample the first neutron interactions. To overcome the large statistical errors of the inelastic gamma-ray scores, the neutron inelastic interaction is always forced to occur at first, and then another interaction is sampled between the elastic scattering interaction and the radiative capture interaction. The tracked neutron is split into two new neutrons: one having the inelastic scattering interaction and the other having either the elastic scattering interaction or the radiative capture interaction. The two new neutrons are tracked independently later. The second technique is applied to sample the interaction type in the sample region. If the total number of the neutron radiative capture interactions is less than a maximum value preset in the code, the neutron radiative capture interaction is forced to take place first, and then another interaction is sampled between the elastic scattering interaction and the inelastic scattering interaction. The third technique is applied to the remaining situations. In this case, the interaction type is sampled among the neutron inelastic scattering, neutron elastic scattering, and neutron radiative capture interactions simply based on their probabilities.

After an elastic scattering interaction and the element with which the neutron has an interaction are sampled, the new flight direction of the neutron and its energy after the neutron inelastic scattering need to be determined. The Monte Carlo models used in the current code depend on the incident neutron kinetic energy and the target nucleus. If the incident neutron energy is within the thermal energy range, the free gas kernel[22] is employed since it results in a good approximation to the thermal flux spectrum and can be sampled without the use of tables. For those incident neutrons in the rest of the energy region, if the target nucleus has a large atomic number such as lead, the anisotropic neutron scattering model is used; otherwise, the isotropic neutron scattering model is selected. When the incident neutron kinetic energy is high enough to drive the target nucleus into its excited state, the inelastic scattering reaction usually has a high probability. Once the inelastic scattering type and the element are determined, the exiting neutron energy and new direction are then sampled.

The Monte Carlo simulations for neutron transport are nonanalog. The accumulated neutron weight has to be adjusted whenever any fictitious probability density function is sampled. In addition, if it falls below the neutron weight cutoff value, Russian roulette is played to determine if the neutron will be killed or tracked with an increased weight.

IV. GAMMA-RAY TRANSPORT

The gamma-ray cross-section data were extracted from the ENDF/B-VI Release 8 EPDL library and were

processed into three separate tables, which are total, Compton scattering, and pair production cross-section tables. These tables cover the elements from $Z = 1$ to 100 and the energy range from 10 keV to 20 MeV.

The CEARPGA II code models the transport of all the gamma rays from

1. neutron radiative capture interactions
2. neutron inelastic interactions
3. neutron source fission reactions
4. radioisotope decays
5. backgrounds including natural backgrounds, NaI detector activations, and gamma rays from the structural materials and the neutron source.

These gamma rays are treated by three different approaches:

1. analog Monte Carlo simulation
2. analog linear interpolation
3. sampling deposited gamma-ray energy.

IV.A. The Analog Linear Interpolation Approach

This approach is applied specifically to gamma rays generated from neutron radiative capture interactions, radioisotope decays in the sample region, and the natural background. It is a combination of the Analog Monte Carlo simulation method and the linear interpolation technique. With this approach, once the tracked neutron enters the sample region and the radiative capture interaction is chosen, only the pseudo gamma rays are forced to be emitted and are tracked separately by an analog Monte Carlo approach. The implementation of the analog linear interpolation is described as follows.

IV.A.1. Choosing Proper Pseudo Gamma Rays

A set of pseudo gamma rays is selected to represent those gamma rays generated from neutron radiative capture interactions, radioisotope decays in the sample region, and the natural background. The only requirement is that they should span the energy range of the actual gamma rays so that interpolation can be used instead of extrapolation. In the current code, 24 pseudo gamma rays are used, with energies of 0.5, 1.0, 1.5, 2.0,…,12 MeV respectively. The initial weight for each pseudo gamma ray is given by

$$W = \frac{W_0}{\Sigma_t} \quad , \tag{5}$$

where W_0 is the cumulative weight of the neutron from its origin to the present position where the radiative capture interaction is sampled and Σ_t is the total radiative capture cross section of the sample material, given the incident neutron energy.

IV.A.2. Optimizing the Gamma-Ray Sampling Number

In order to reduce the statistical error, each pseudo gamma ray is tracked a number of times independently starting from its origin before moving to track the next pseudo gamma ray. The selection of the gamma-ray sampling number (or tracking number) is a trade-off between increasing the gamma-ray scoring from each site and avoiding spending too much time on each gamma ray, which may result in a decrease of the entire calculation efficiency. Trial and error is usually a good way to optimize the gamma-ray sampling number. In the CEARPGA II code, the default gamma-ray sampling number is set to be 100.

IV.A.3. Using the Analog Monte Carlo Method for Tracking Pseudo Gamma Rays

Each pseudo gamma ray is tracked basically using the analog Monte Carlo method. The only variance reduction method used is implicit capture.

IV.A.4. Tallying the Scores of Pseudo Gamma Rays

The score of an incident gamma ray is the product of its weight accumulated along the track to the detector and detector efficiency, where the detector efficiency is defined as the ratio of actual recorded number of gamma rays to the total number of gamma rays that are incident on the detector and is calculated by

$$\varepsilon = 1 - e^{-(\Sigma_t \cdot l)} \quad , \tag{6}$$

where Σ_t is the total cross section of the incident gamma ray in the detector and l is the intersection length of the incident gamma-ray flight path with the detector.

All scores of incident gamma rays are recorded to a set of energy-score tables of two dimensions, one representing the incident gamma-ray energy expressed in terms of the channel number and the other representing the scores of the incident gamma rays that originate from the same pseudo gamma ray of interest. The energy channel N for the incident gamma ray of energy E_f is determined by the following formula:

$$(N-1)\cdot\Delta E < E_f \le N\cdot\Delta E \quad , \tag{7}$$

where ΔE is the energy increment between two consecutive channels.

When the pseudo gamma ray has a pair production interaction outside the detector and generates two 0.511-MeV annihilation gamma rays, its score cannot be recorded until the tracking of both 0.511-MeV gamma rays is finished. Assuming that the energy of the corresponding pseudo gamma ray is E_i, the generated pair production

gamma rays have incident energies of E_{p1} and E_{p2} and corresponding scores of W_{p1} and W_{p2}, where $W_{p1} \geq 0$ and $W_{p2} \geq 0$. The detailed tally procedure is described as follows:

1. Add the score of $W_{p1} \times W_{p2}$ to the table of the pseudo gamma ray E_i with the energy channel determined by incident energy $E_{p1} + E_{p2}$.

2. Add the score of $W_{p1} - W_{p1} \times W_{p2}$ to the table of the pseudo gamma ray E_i with the energy channel determined by incident energy E_{p1}.

3. Add the score of $W_2 - W_{p1} \times W_{p2}$ to the table of the pseudo gamma ray E_i with the energy channel determined by incident energy E_{p2}.

IV.A.5. Calculating the Average Neutron Capture Cross Section of Each Element in the Sample Material

For each element in the sample, the average neutron capture macroscopic cross section is calculated during the process of simulation and is used to adjust the interpolated incident gamma-ray spectrum later. For the i'th element in the sample region, the average cross section $\mu^i_{(n,r)}$ is calculated according to the following formula:

$$\mu^i_{(n,r)} = \frac{\sum_j w^i_j \mu^i_j}{\sum_j w^i_j} \quad , \tag{8}$$

where

μ^i_j = neutron radiative capture macroscopic cross section of the element i for the j'th neutron capture interaction in the sample

w^i_j = weight factor of the neutron in the j'th neutron capture interaction.

IV.A.6. Implementing the Interpolation

Once a set of energy-score tables is established, linear interpolation can be used to derive the incident spectrum for any gamma ray generated in the sample region. Given M pseudo prompt gamma rays with energies of $E_0, E_1, E_2, \ldots, E_{M-1} (E_0 < E_1 < E_2 < \cdots < E_{M-1})$ and their corresponding energy-score tables $T_0, T_1, T_2, \ldots, T_{M-1}$ with maximum channel numbers of $N_0^{max}, N_1^{max}, \ldots, N_{M-1}^{max}$, respectively, the interpolation procedure is described below.

Choosing the Appropriate Energy-Score Tables

If the incident gamma-ray energy E is found to be located between E_{k-1} and E_k, where $k \leq M-1$, then the energy-score tables T_{k-1} and T_k are to be used for interpolation. As an exception, if E is found $< E_1$, the first two tables (T_1 and T_2) are chosen.

Adjusting the Selected Energy-Score Tables

The selected energy-score tables are then adjusted in a way that their scores are redistributed within the same channels whose range is determined by the interpolated incident gamma-ray energy. Assuming N^{max}, N_{in}^{max} are the full energy channel numbers for the selected pseudo-gamma-ray energy-score table and the incident gamma ray, the adjusting procedure for the selected pseudo-gamma-ray energy-score table is described as follows:

1. Assign the score in the full energy channel N^{max} to the channel N_{in}^{max}.

2. Assign the total scores between channels $(K-1)(N^{max}-1)/(N_{in}^{max}-1)$ and $K(N^{max}-1)/(N_{in}^{max}-1)$ of the selected energy-score table to the channel K of the modified energy-score table ($K = 1,2,\ldots,N_{in}^{max}-1$). Note that when N^{max} is $> N_{in}^{max}$, the selected energy-score table will be compressed into the modified energy-score table; on the contrary, when N^{max} is smaller than N_{in}^{max}, the selected energy-score table will be expanded into the modified energy-score table. In both cases, linear interpolation is always used.

Deriving the Incident Spectrum for the Incident Gamma Ray

The scores of the incident gamma ray with unit weight are interpolated by using two adjusted energy-score tables, as given by

$$S^i = \frac{\lfloor (\ln E - \ln E_{k-1}) \cdot S^i_K + (\ln E_K - \ln E) \cdot S^i_{K-1} \rfloor}{\ln E_k - \ln E_{k-1}} \quad , \tag{9}$$

where

S^i = interpolated score for energy channel i of the gamma ray of concern

E = gamma-ray energy (MeV)

E_{K-1} = energy of the $(K-1)$'th pseudo gamma ray (MeV)

S^i_{K-1} = score in channel i of the modified energy-score table for the selected $(K-1)$'th pseudo gamma ray

E_K = energy of the K'th pseudo gamma ray (MeV)

S^i_K = score in channel i of the modified energy-score table for the selected K'th pseudo gamma ray.

With this approach, all elemental library spectra (including prompt-gamma-ray and decay-gamma-ray contributions) and three natural background libraries from the ^{40}K decay chain, the thorium decay chain, and the uranium decay chains, respectively, can be obtained.

IV.A.7. Generating the Spectra for Prompt Gamma Rays

Once the energy-score tables are established, the linear interpolation approach is applied to derive the incident prompt-gamma-ray spectra and decay-gamma-ray spectra of the sample material and natural backgrounds. This procedure is described as follows:

1. Select the element of interest.

2. Determine the gamma-ray energy and its yield that needs to be interpolated.

3. For each gamma ray, interpolate the energy-score tables to obtain its corresponding incident spectrum.

4. Modify the incident spectrum by multiplying appropriate adjusting factors.

For prompt gamma rays, there are two adjusting factors: gamma-ray yield and elemental average neutron absorption cross section obtained during the simulation. For decay gamma rays, in addition to the above two factors, there is still another factor: the expected value of the decay chance δ, which is given by[16]

$$\delta = \left[1 - \frac{(1 - e^{-\lambda t_0})}{\lambda t_0} \right] , \tag{10}$$

where

$\lambda = \ln 2/T$ = decay constant for a certain radioisotope

t_0 = measurement time (s).

For gamma rays from the natural backgrounds—the ^{40}K decay chain, thorium decay chain, and uranium decay chain in the sample—the corresponding relative intensity is the only factor.

5. Add the interpolated incident spectrum to the total incident gamma-ray spectrum of the element of interest.

6. Repeat steps 2 through 5 until the interpolations for all the gamma rays associated with the element have been completed.

IV.B. The Analog Monte Carlo Approach

This approach is applied specifically to the gamma rays from the neutron source fission reaction, inelastic scattering interactions, and radiative capture interactions in the regions excluding the sample region. Whenever gamma rays from these sources are generated, they are tracked independently in an analog Monte Carlo way. The implementation of this approach is simple. The sampling schemes for gamma-ray transport including flight direction, flight distance to the next interaction site, interaction type, new energy after interaction, etc., are well described in the references.[22,23] To increase the scoring chance at each interaction site, each gamma ray is always tracked a number of times independently, as in the treatment of the pseudo gamma rays. Considering that the photoelectric interaction of gamma rays can only lead to the loss of gamma rays, it is always treated implicitly. A weight-adjusting factor is assigned to modify the gamma-ray weight after sampling the interaction, as given by

$$W = \frac{\Sigma_{pp} + \Sigma_C}{\Sigma_t} , \tag{11}$$

where

Σ_t = total macroscopic cross section in the material

Σ_{pp} = macroscopic pair production cross section

Σ_C = macroscopic Compton cross section.

Neutron source gamma rays are tabulated in 18 energy groups over the energy range from 0 to 7.2 MeV based on the ^{252}Cf fission gamma-ray energy spectrum.[24] When a new neutron history starts, 18 gamma rays are forced to be emitted (one gamma ray from each energy group). The scores of these gamma rays are tallied into the neutron source library.

The inelastic gamma-ray data are extracted from the reference.[25] The generation of the inelastic gamma rays depends on the sampled excited level for the selected element and the incident neutron energy once an inelastic scattering interaction is sampled. The scores of inelastic gamma rays from the regions other than the sample region are tallied to the "Extras" library, while the scores of inelastic gamma rays from the sample region are tallied to the corresponding elemental library spectra.

Prompt gamma rays generated from the radiative capture interactions outside the sample region are always forced to be emitted once a radiative capture interaction is sampled. The scores of these gamma rays are tallied into the "Extras" library.

Decay gamma rays emitted from regions other than the sample region are forced to be emitted along with the prompt gamma rays and are treated just like the prompt gamma rays with the exception that the expected value of the decay chance δ is used to modify each decay-gamma-ray yield. The scores of these gamma rays are tallied into the "Extras" library as well.

IV.C. Sampling the Deposited Gamma-Ray Energy

This approach is used specifically to the NaI detector activation gamma rays including the prompt gamma rays from I and Na and decay gamma rays from the radioisotopes ^{128}I and ^{24}Na. It is based on the assumption that the deposited energy of a gamma ray inside the detector can be considered as a random variable with the

probability density distribution equal to its detector response function. Therefore, the deposited gamma-ray energy can be sampled according to the detector response function. The implementation of this approach is described as follows:

1. Force the radiative capture interactions with both Na and I to take place once a neutron enters the NaI detector.

2. Force all the coincident prompt gamma rays and decay gamma rays to be emitted after capture interaction.

3. Find the first interaction position of each coincident gamma ray. This requires sampling the initial direction and flight path length to the first interaction site of each coincident gamma ray to determine whether the gamma ray escapes from the detector or not.

4. Determine the deposited energy of the coincident gamma ray. Assume $E_{i,j}^{k}$ is the gamma-ray energy for the i'th coincident gamma ray in the j'th coincident scheme for the k'th element; $f(E)$ is the detector response function for the incident gamma ray with the energy equal to $E_{i,j}^{k}$, and the actually deposited gamma-ray energy in mega-electron-volts is determined according to the following rules. If the gamma ray escapes out of the NaI detector, the deposited energy $E_{i,j,k}^{D} = 0$. If the first interaction of the gamma ray is inside the detector, the deposited gamma-ray energy is given by

$$\int_{0}^{E_{i,j,k}^{D}} f(E)\,dE \geq \xi \quad , \tag{12}$$

where ξ is the sampled random number between 0 and 1.

5. Determine the energy of the coincident beta particle. For the decays of ^{24}Na and ^{128}I, most decay schemes involve the generation of a beta particle with a certain characteristic maximum energy in addition to gamma rays that are coincident with it. In this case, the detected energy is the sum of the deposited energies of all coincident gamma rays and the beta particle. The energy distributions for the beta particle accompanying the ^{24}Na decay scheme is from Kantele,[26] while the energy distribution for the beta particle accompanying the ^{128}I decay scheme is an approximation based on ^{114}In decay due to the lack of data.[19] These energy distributions of beta particles for both radioisotopes were fitted to a third-order polynomial. Both are then normalized as a probability density function by the following:

$$f(E) = \frac{a_1 E^3 + a_2 E^2 + a_3 E + a_4}{\int_{0}^{E_{endpoint}} (a_1 E^3 + a_2 E^2 + a_3 E + a_4)\,dE} \quad , \tag{13}$$

where a_1 through a_4 are fitting coefficients.

The deposited energy of the beta particle is obtained by sampling the fitted probability density distributions for beta energy distributions using the rejection technique.

6. Tally the scores for prompt gamma rays and decay gamma rays. The scores of prompt gamma rays are tallied to the prompt-gamma-ray libraries of both I and Na. The tallied energy is the sum of the energy deposited by each coincident gamma ray, represented by

$$E_j = \sum_{i=0}^{N} A \cdot E_{i,j,k}^{D} \quad , \tag{14}$$

where $A = 0$ if the coincident gamma ray escapes out of the NaI detector; otherwise, $A = 1$. N is the total number of coincident gamma rays in the j'th coincident scheme. The tallied score is given by

$$W = W_0 \cdot \frac{\Sigma_k^a}{\Sigma_t} \cdot \frac{\eta_k^j}{\eta_k} \quad , \tag{15}$$

where

W_0 = cumulative neutron weight at the entrance of the NaI detector

Σ_t = total cross section of the neutron interaction for the NaI detector

Σ_a^k = total radiative capture interaction cross section for the element k in the NaI detector; k is either I or Na

η_k^j = intensity of the coincidence scheme j for element k.

The scores of decay gamma rays from radioisotopes ^{128}I and ^{24}Na are tallied to the ^{128}I decay library and ^{24}Na decay library, respectively. The tallied energy is determined by

$$E_j = \sum_{i=0}^{N} A \cdot E_{j,D}^{i} + E_\beta \quad , \tag{16}$$

where E_β is the sampled beta particle energy in a scheme.

The tallied score has the same description as that for prompt gamma rays as mentioned before.

V. BENCHMARKING THE CEARPGA II CODE

The CEARPGA II code has been benchmarked against the experimental data, and the simulated results have been obtained from the CEARPGA I code and the MCNP4C code.

Six coal sample spectra provided by Energy Technologies, Inc., were obtained from a PGNAA coal analyzer prototype and are shown in Fig. 2. The sulfur amounts in these six coal samples are given in Table I. Each coal spectrum was accumulated for 5 h with a 2.9-μg

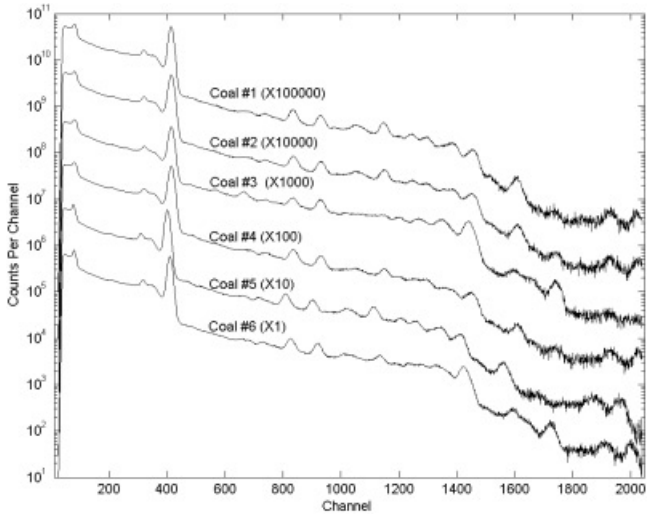

Fig. 2. Six experimental coal spectra before calibration.

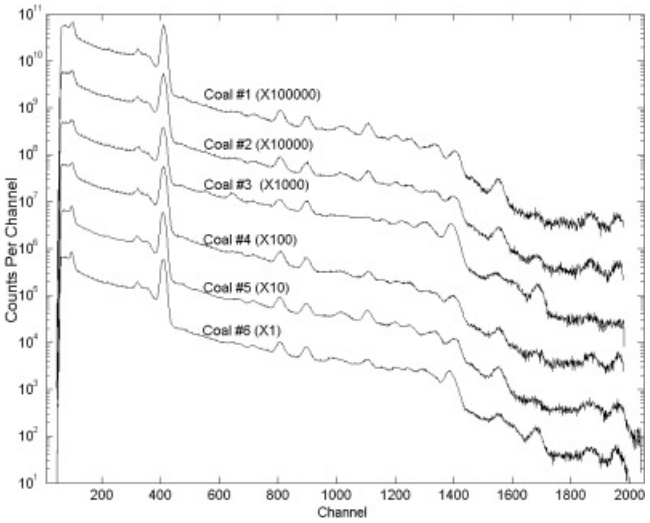

Fig. 3. Six coal experimental spectra after calibration.

^{252}Cf source. It is clear that these measured six spectra are not at the same zero and gain calibration. Therefore, for the purpose of the application and comparison, they must be calibrated. Figure 3 shows the calibrated coal spectra that were shifted with the GZNAI3 code by putting the same and easily identified peaks of the spectra such as the 2.223-MeV hydrogen and 10.829-MeV nitrogen peak, etc., in the same channels of the simulated coal spectrum as those from the CEARPGA II code.

The Monte Carlo model for the coal analyzer geometry is schematically shown in Fig. 4, which consists of a ^{252}Cf radioisotope neutron source, a 6×6 NaI detector, and a rectangular coal chute located between the source and the detector. The cylindrical analyzer body is filled with neutron moderators and shielding materials such as polyethylene, paraffin, lead, etc. In the Monte Carlo simulations, the average compositions of the six coal samples were used and are shown in Table II. The coal density was assumed to be 0.86 g/cm^3, and the neutron source strength was set to be 6.8434×10^6 n/s. The numbers of simulation histories were 200 000 for the CEARPGA II code and 2 million for the CEARPGA I code.

The CEARPGA II–simulated coal sample spectrum is shown in Fig. 5 with the CEARPGA I–simulated spectrum and the two experimental spectra for samples 4 and 6. Obviously, no big weight peaks are observed in the CEARPGA II–simulated spectrum while they are

TABLE I

Sulfur Experimental and MCLLS Amounts in Six Coals Used

Coal	Sulfur Amount (%)	
	Experimentally Measured	MCLLS Calculated
1	0.57	0.66
2	0.68	1.20
3	0.36	0.33
4	0.78	0.61
5	0.61	0.75
6	1.19	0.56

TABLE II

Elemental Compositions for Coal Sample Used for Monte Carlo Simulation

Element	Weight Fraction (%)
Hydrogen	5.20
Carbon	75.2865
Nitrogen	1.42
Oxygen	12.5516
Sodium	0.045
Magnesium	0.0595
Aluminum	1.164
Silicon	2.2612
Phosphorous	0.0097
Sulfur	0.77
Chlorine	0.11
Potassium	0.3277
Calcium	0.108
Titanium	0.0656
Manganese	0.0001
Iron	0.6212
Nickel	0.0001

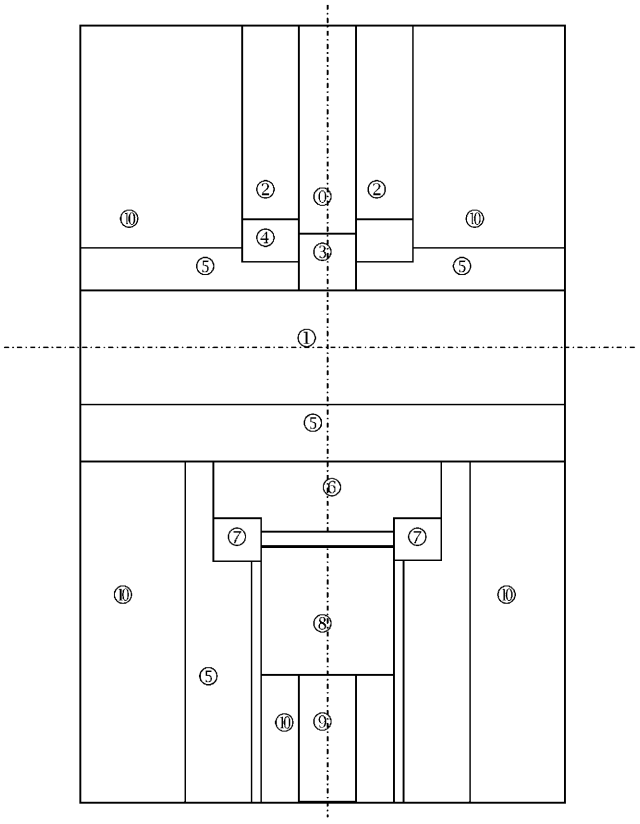

Fig. 4. Schematic of prototype coal analyzer model: ① coal sample, ② polyethylene, ③ bismuth with neutron source, ④ air, ⑤ aluminum, ⑥ lithium-polyethylene, ⑦ lead, ⑧ NaI detector, ⑨ SiO_2 (PMT), and ⑩ paraffin.

observed in the CEARPGA I–simulated spectrum around channels 226, 185, and 160. The CEARPGA II simulation also shows very good agreement with the measured spectra with respect to the corresponding energy peak positions and their heights as well as the overall spectral shape. However, the magnitude of the counts in most channels of the CEARPGA II spectrum is about two times the measured spectra with the exception of the part below channel 420, which is about four times those of the measured spectra. This phenomenon of the overall spectral shift upward was also observed in the CEARPGA I–simulated spectrum, where the calculated counts per channel is nearly 20 times as large as that in the measured spectra though the simulations were run based on the same conditions. There are several factors that may be responsible for the shift:

1. differences in the sample composition used for the simulation and the actual experimental values
2. differences in the geometrical model and the actual geometry configuration

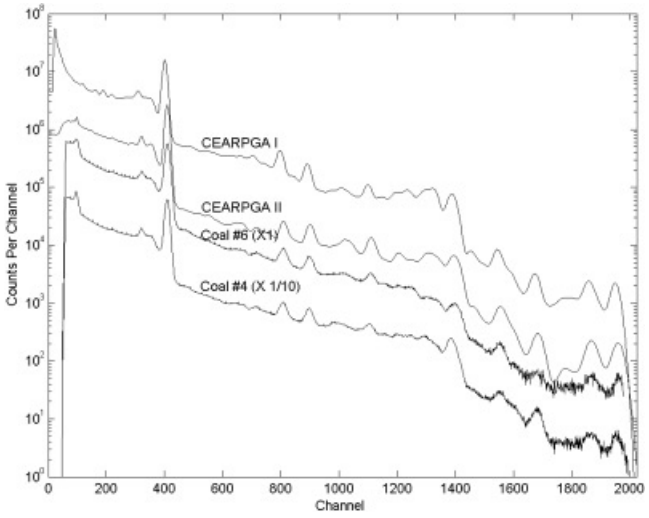

Fig. 5. Simulated coal sample spectra from CEARPGA I and II codes and their comparison with experimental spectra (coal #4 and coal #6).

3. differences in the structural material compositions
4. some possible bugs in the code that are currently unknown.

To determine the accuracy of the CEARPGA II code, its simulated spectrum was benchmarked against the simulated results of the well-accepted MCNP code, and the results are shown in Fig. 6. The number of simulation histories for the MCNP code was 20 million without use of any variance reduction techniques. The MCNP-calculated pulse-height tallies were Gaussian spread by

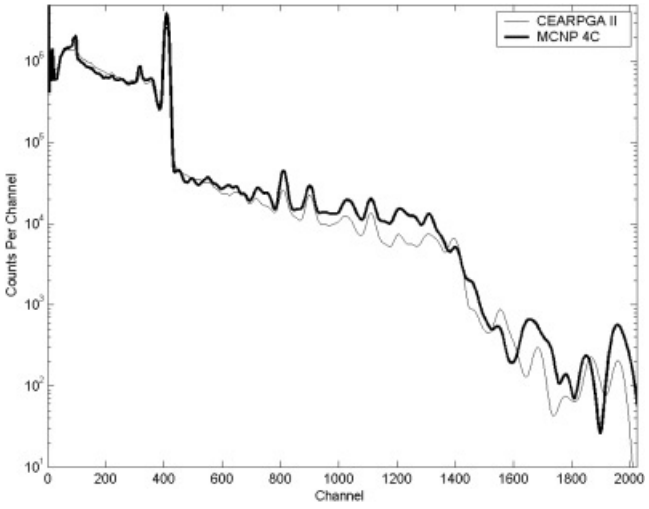

Fig. 6. CEARPGA II code and MCNP4C-simulated coal sample spectra.

using the same program developed for the CEARPGA codes. It is clear that both spectra exhibit similar energy peaks over the whole range. Especially in the lower-energy range up to channel 450, the two spectra match very well, and no obvious shift is observed. Therefore, it can be concluded that the CEARPGA II code itself has nothing to do with the intensity shift. As far as those channels greater than 450, the calculated results by CEARPGA II are relatively smaller than those calculated by the MCNP code. When the whole spectral peak shapes are taken into account and compared to the experimental spectra, CEARPGA II seems to give a better result than MCNP does. This is probably primarily due to the lack of statistical significance in the MCNP-calculated spectrum.

The CEARPGA II–calculated three background libraries from neutron activation of the NaI detector were benchmarked against the corresponding experimentally determined library spectra obtained by Gardner et al.[14] As shown in Figs. 7, 8, and 9, the ^{24}Na and ^{128}I spectra of radioisotope decay beta particles in coincidence with gamma rays are consistent with their experimental spectra. But, the simulated spectrum for prompt gamma rays does not agree so well with the experimental spectrum, especially in the lower-energy range. This is to be expected because at the present time the complete data for the coincidence schemes for the prompt gamma rays of Na and I are not available.

Library least-squares analysis was performed for the six experimental coal spectra using the set of library spectra that were calculated from the CEARPGA II code. The calculated sulfur amounts in six coal samples are listed in Table I along with the corresponding experimental sulfur amount. As an example, the library least-squares–fitted spectrum for coal 1 is compared to its experimental spectrum and is given in Fig. 10.

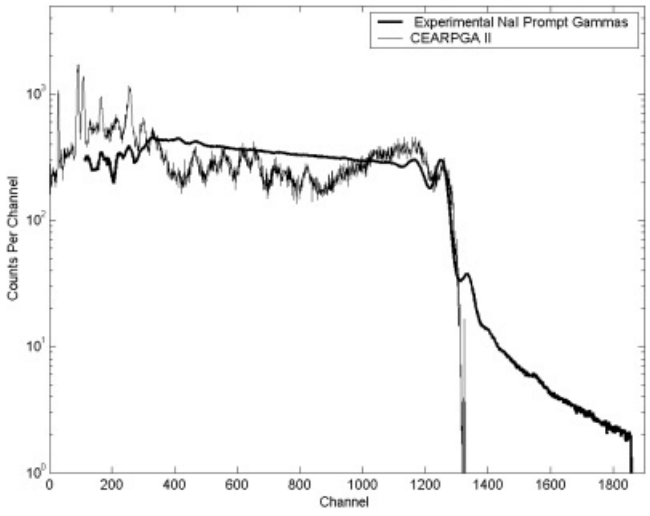

Fig. 7. CEARPGA II–simulated prompt-gamma-ray spectrum from neutron activation of NaI detector and experimentally measured spectrum.

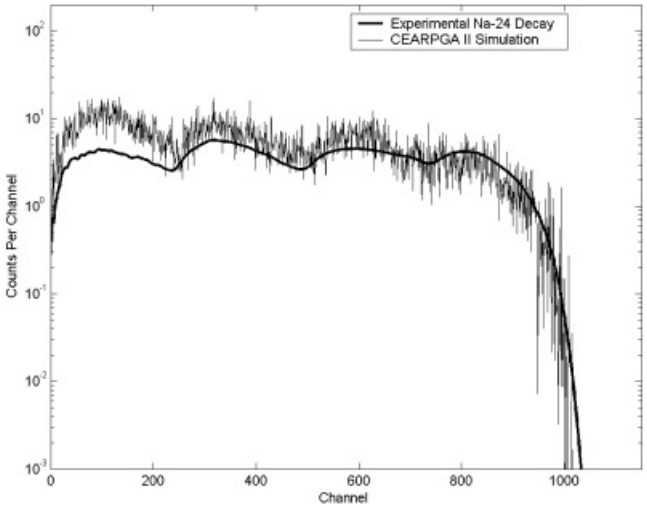

Fig. 8. CEARPGA II–simulated ^{24}Na decay spectrum from neutron activation of NaI detector and experimentally measured spectrum.

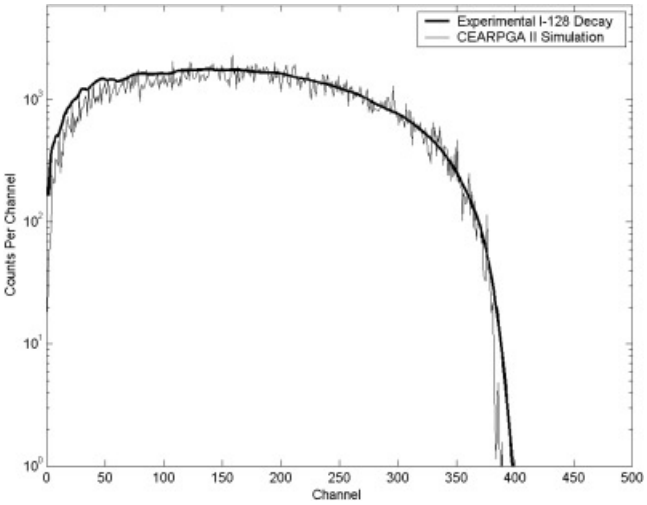

Fig. 9. CEARPGA II–simulated ^{128}I decay spectrum from neutron activation of NaI detector and experimentally measured spectrum.

VI. DISCUSSION AND CONCLUSIONS

As a Monte Carlo simulation code for the PGNAA analysis, the CEARPGA II code not only calculates the total pulse-height spectrum from the characteristic gamma rays but also generates a set of libraries required for the MCLLS analysis, including all elemental library spectra

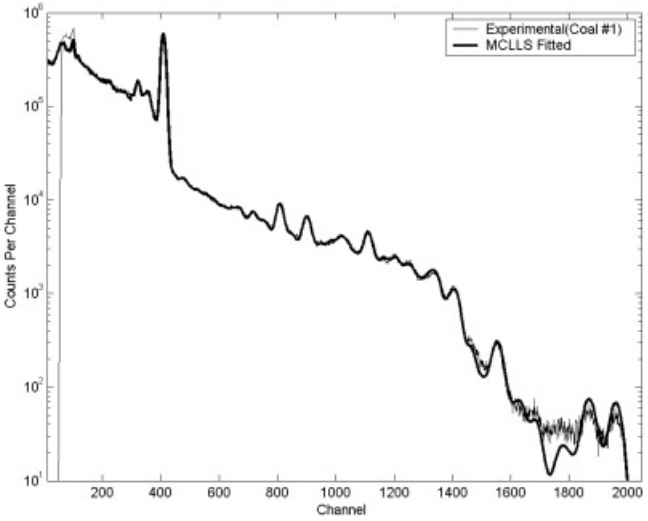

Fig. 10. Experimental spectrum and MCLLS fitted spectrum for coal #1.

in the sample, one background library generated for the gamma rays of the neutron source, one for the structural material surrounding the sample, three background libraries generated for the NaI detector activation spectra, and three natural background libraries.

Compared to its previous version, several important improvements have been made. They are

1. tracking all the annihilation gamma rays from the pair production interactions outside the detector
2. simulating the neutron activation background spectra
3. simulating the natural background spectra
4. accounting for the nonlinearity and flat continuum of the NaI detectors
5. adopting a general geometry package
6. solving the big weight problem.

The simulated result from the CEARPGA II code has been benchmarked against those calculated from the CEARPGA I code and the MCNP code and from the experimental sample spectra. Results demonstrate that the big weight problem observed from the spectrum calculated by the CEARPGA I code has been successfully eliminated in the CEARPGA II code. In addition, the CEARGPA II code shows improved calculational efficiency.

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REFERENCES

1. C. SHYU, "Development of the Monte Carlo–Library Least-Squares Method of Analysis for Neutron Capture Prompt Gamma-Ray Analyzer," PhD Dissertation, North Carolina State University (1991).

2. C. M. SHYU, R. P. GARDNER, and K. VERGHESE, "Development of the Monte Carlo–Library Least-Squares Method of Analysis for Neutron Capture Prompt Gamma-Ray Analyzers," *Nucl. Geophys.*, **7**, *2*, 241 (1993).

3. R. P. GARDNER, "Software Design for a PGNAA Coal Analyzer Prototype," North Carolina State University (2000).

4. C. D. ZERBY, A. MEYER, and R. B. MURRAY, "Intrinsic Line Broadening in NaI(Tl) Gamma-Ray Spectrometers," *Nucl. Instrum. Methods*, **12**, 115 (1961).

5. W. C. KAISER, S. I. BAKER, A. J. MACKAY, and I. S. SHERMAN, "Response of NaI (Tl) to X-Rays and Low Energy Gamma Rays," *IRE Trans. Nucl. Sci.*, **NS-9**, *3*, 22 (1962).

6. A. J. L. COLLINSON and R. HILL, "The Fluorescent Response of NaI(Tl) and CsI(Tl) to X Rays and γ Rays," *Proc. Phys. Soc.*, **81**, 883 (1963).

7. R. L. HEATH, "Scintillation Spectrometry Gamma-Ray Spectrum Catalogue," IDO-16880-1, AEC Research and Development Report, Physics, TID-4500 (1964).

8. J. D. VALENTINE and B. D. ROONEY, "Design of a Compton Spectrometer Experiment for Studying Scintillator Nonlinearity and Intrinsic Energy Resolution," *Nucl. Instrum. Methods A*, **353**, 37 (1994).

9. B. D. ROONEY and J. D. VALENTINE, "Scintillator Light Yield Nonproportionality: Calculating Photon Response Using Measured Electron Response," *IEEE Trans. Nucl. Sci.*, **43**, *3*, 1271 (1997).

10. J. D. VALENTINE, B. D. ROONEY, and P. DORENBOS, "More on the Scintillation Response of NaI(Tl)," *IEEE Trans. Nucl. Sci.*, **45**, *3*, 1750 (1998).

11. J. D. VALENTINE, B. D. ROONEY, and J. LI, "The Light Yield Nonproportionality Component of Scintillator Energy Resolution," *IEEE Trans. Nucl. Sci.*, **45**, *3*, 512 (1998).

12. R. P. GARDNER and C. W. MAYO, "NaI Detector Nonlinearity for PGNAA Applications," *Appl. Rad. Isot.*, **51**, 189 (1999).

13. R. P. GARDNER and A. SOOD, "A Monte Carlo Simulation Approach for Generating NaI Detector Response Functions (DRF's) that Accounts for Nonlinearity and Variable Flat Continua," *Nucl. Instrum. Methods B*, **213**, 87 (2004).

14. R. P. GARDNER, E. SAYYED, Y. ZHENG, S. HAYDEN, and C. W. MAYO, "NaI Detector Neutron Activation Spectra for PGNAA Applications," *Appl. Radiat. Isot.*, **53**, 483 (2000).

15. A. SOOD, "A New Monte Carlo Assisted Approach to Detector Response Functions," PhD Thesis, North Carolina State University (2000).

16. W. ZHANG, "The Analog Linear Interpolation Approach for Monte Carlo Simulation of Prompt Gamma-Ray Neutron Activation Analysis," PhD Thesis, North Carolina State University (2003).

17. W. ZHANG and R. P. GARDNER, "The Analog Linear Interpolation Approach for Monte Carlo Simulation of PGNAA: The CEARPGA Code," *Nucl. Instrum. Methods B*, **213**, 116 (2004).

18. R. P. GARDNER, A. M. YACOUT, J. ZHANG, and K. VERGHESE, "An Investigation of the Possible Interaction Mechanisms for SiLi and Ge Detector Response Functions by Monte Carlo Simulation," *Nucl. Instrum. Methods Phys. Res. A*, **242**, 399 (1986).

19. E. EL-SAYYED, "NaI Detector Neutron Activation Spectra for PGNAA Applications," MS Thesis (2000).

20. Y. L. YUAN, R. P. GARDNER, and K. VERGHESE, "A Monte Carlo Model for On-Line Neutron Capture Prompt Gamma-Ray Analysis of Coal in Transmission Geometry," *Nucl. Technol.*, **77**, 97 (1987).

21. J. R. BEYSTER, H. ANTUNEZ, and W. BROUWER, "Integral Neutron Thermalization," GA-6824, General Atomics Company (1965).

22. L. L. CARTER and E. D. CASHWELL, "Particle-Transport Simulation with the Monte Carlo Method," TID-26607, U.S. Department of Energy (1975).

23. "MCNP4C," CCC-700, U.S. Department of Energy (2000).

24. D. T. INGERSOL and B. W. WEHRING, "Gamma-Ray Pulse-Height Response of an Ne-213 Scintillation Detector," *Nucl. Instrum. Methods*, **147**, 55 (1977).

25. M. C. BERTIN, J. CELINIK, H. FLESHER, G. GROCHOWSKI, M. H. KALOS, J. H. RAY, and E. S. TROUBETZKOY, UNC-5099, United Nuclear Corporation (1964).

26. J. KANTELE, *Handbook of Nuclear Spectrometry*, Academic Press, San Diego, California (1995).