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The analog linear interpolation approach for Monte Carlo simulation of PGNAA: The CEARPGA code

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Abstract

The analog linear interpolation approach (ALI) has been developed and implemented to eliminate the big weight problem in the Monte Carlo simulation code CEARPGA. The CEARPGA code was previously developed to generate elemental library spectra for using the Monte Carlo – library least-squares (MCLLS) approach in prompt gamma-ray neutron activation analysis (PGNAA). In addition, some other improvements to this code have been introduced, including (1) adopting the latest photon cross-section data, (2) using an improved detector response function, (3) adding the neutron activation backgrounds, (4) generating the individual natural background libraries, (5) adding the tracking of annihilation photons from pair production interactions outside of the detector and (6) adopting a general geometry package. The simulated result from the new CEARPGA code is compared with those calculated from the previous CEARPGA code and the MCNP code and experimental data. The new CEARPGA code is found to give the best result.

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1. Introduction

The Monte Carlo – library least-squares (MCLLS) method [3,8] was proposed to analyze the contents of sample material in prompt gamma-ray neutron activation analysis (PGNAA). This approach takes advantage of Monte Carlo simulation to generate the required elemental library spectra for the sample of interest as opposed to using experimental methods, thereby saving a lot of tedious and costly experimental work.

The CEARPGA code, developed in the Center for Engineering Applications of Radioisotopes of North Carolina State University, was used to implement the MCLLS algorithm for coal sample analysis [7,8]. The code incorporated a variety of variance reduction techniques to improve calculation efficiency such as forcing all prompt gamma rays to be emitted after a neutron interaction (stratified sampling), using the expected value splitting (EVS) technique to increase the score probability of each tracked gamma ray, using the correlated sampling method to deal with small variations of sample compositions, and using a

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detector response function to convert the incident gamma-ray spectra on the detector to pulse height spectra. While this code is efficient and accurate, it suffers from the big weight problem in which a few histories yield very large weights with very large variances. This big weight problem was found to be caused primarily by the expected value splitting technique used for tracking gamma rays [7].

In order to minimize or eliminate this problem while still maintaining high calculational efficiency, the authors have investigated two approaches: (1) the score importance map approach combined with the batch tracking technique and (2) the analog linear interpolation approach (ALI). Initial studies show that both approaches are effective, but the ALI is judged to be better and has been implemented in the new CEARPGA code.

In this paper, the ALI approach is described along with some other important improvements to the CEARPGA code and the calculated results are compared with those calculated by the previous CEARPGA code, the MCNP (4C) code and from measured data.

2. The analog linear interpolation approach

The ALI approach [9] is a combination of the analog Monte Carlo simulation method and a linear interpolation technique and is mainly employed to track prompt gamma rays emitted from neutron capture interactions in the sample. In this approach a set of pseudo gamma rays that represent the actual prompt gamma rays are first tracked by analog Monte Carlo to establish a set of energy-score tables. Later the linear interpolation approach is used to derive any incident prompt gamma-ray spectrum of interest, based on the energy-score tables. The procedure of implementing the ALI approach consists of the following steps:

(1) Choosing the number and energy of the pseudo gamma rays.

Instead of tracking all prompt gamma rays emitted from the neutron capture interactions in the sample of interest, only a set of pseudo gamma rays are selected and tracked as representatives of all the prompt gamma rays. The pseudo gamma rays should cover the energy range of the actual prompt gamma rays so that interpolation can be used later.

(2) Determining the number of pseudo gamma rays to sample.

The number of pseudo gamma rays to sample of each energy is determined by trial and error to give optimum statistics.

(3) Tracking the pseudo gamma rays.

Each pseudo gamma ray is tracked starting from the site of the current neutron radiative capture interaction by the analog Monte Carlo approach, except that the photoelectric absorption is sampled implicitly.

(4) Tallying the scores of pseudo gamma rays.

The magnitude of the score that a pseudo gamma ray has is the product of its weight accumulated along its track to the detector and the detector efficiency. The scores of incident gamma rays are recorded to a set of energy-score tables based on both the pseudo gamma-ray energy and the incident energy. The energy-score tables are of two dimensions, with one dimension representing the energy of incident gamma rays and the other representing the total scores of incident gamma rays in the previous histories. The score scheme for pair production annihilation photons takes into account the cases that both photons score simultaneously and each independently.

(5) Calculating average neutron capture macroscopic cross-sections.

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

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

where μ^i_j is the neutron radiative capture macroscopic cross-section of element i for the jth neutron capture interaction in the sample,

w_j^i is the weight factor of the neutron in the jth neutron capture interaction.

(6) Interpolating the spectra for all gamma rays of interest.

Incident spectra of gamma rays resulting from neutron capture interactions and radioisotope decays in the sample are obtained through linear interpolation at the end of the complete Monte Carlo simulation after a set of energy-score tables has been established. Note that for any gamma ray of interest, the selected two energy-score tables for interpolation always have different energy ranges. In order to interpolate a new spectrum conveniently, they are converted into two new tables by re-distributing them over the same energy range as determined by the gamma ray to be interpolated.

3. Neutron and gamma-ray transport

Monte Carlo simulation of PGNAA analyzers consists of tracking the transport of both neutrons and associated gamma rays. The sampling schemes for neutron transport have been well documented in the references such as Shyu et al. [8], Shyu [7] and Carter and Cashwell [1] and thus are not described in detail here. Each neutron history starts with sampling the neutron energy and direction from the Cf-252 neutron source and then is tracked inside the analyzer system. To reduce the variance of inelastic scattering spectra, the first interaction of a neutron is forced to be inelastic scattering whenever the inelastic cross-section of the chosen element is greater than zero. The radiative capture interaction is always forced to occur up to 10 times if the neutron sampled has an interaction in the sample region. The neutron of interest is not allowed to escape the system to increase tracking efficiency. Whenever it approaches the system boundary, the truncated exponential probability density function is sampled to make sure the next interaction position is still within the system boundary. The neutron is terminated only if its weight is below a specified cutoff value by the Russian Roulette approach.

Five kinds of gamma-ray sources are considered in the PGNAA simulation. They are neutron radiative capture interaction, neutron inelastic interaction, neutron source fission emission, radioisotope decay, and backgrounds. In the CEARPGA code, gamma rays are further broken into 12 categories: (1) prompt gamma rays from neutron radiative capture interactions in the sample region, (2) decay gamma rays from radioisotopes in the sample region, (3) gamma rays from neutron inelastic scattering in the sample region, (4) prompt gamma rays from neutron radiative capture interactions in regions other than the sample region, (5) gamma rays from neutron inelastic scattering in regions other than the sample region, (6) gamma rays from the neutron source fission reaction, (7) gamma rays from natural background due to the ^{40}K decay chain, (8) gamma rays from natural background due to the Th decay chain, (9) gamma rays from natural background due to the U decay chain, (10) prompt gamma rays from I and Na from the NaI detector neutron activation, (11) decay gamma rays from the ^{24}Na resulting from NaI detector neutron activation, and (12) decay gamma rays from ^{128}I resulting from NaI detector neutron activation.

The gamma rays described are dealt with in different ways. For gamma rays of category 10, they are not modeled directly in the code. The output spectrum at the end of simulation is based on experimentally measured data [4]. For categories 11 and 12, separate Monte Carlo codes are employed [6]. In addition to decay gamma rays, the effect of coincident decay beta particles of ^{24}Na and ^{128}I are also taken into account. For gamma rays of categories 1, 2, 7, 8 and 9, the ALI approach is employed. The rest of the gamma rays are tracked independently. The process of tracking gamma rays is the same as that for pseudo gamma rays with the exception that tallying schemes are different. All the scores of the pseudo gamma rays are tallied to related energy-score tables based on the initial and final energy of the pseudo gamma rays, but the scores of other gamma rays are tallied to their own spectra based only on their final energy. Note that all libraries except that from category 10 are calculated. We are presently working on a Monte Carlo code for calculating this final library, but this must await determination of the coincidence prompt gamma-ray schemes for Na and I.

4. Other CEARPGA code improvements

Apart from the introduction of the ALI approach to solve the big weight problem, several other improvements have also been introduced to the CEARPGA code in order to enhance its simulation accuracy and capabilities. This includes

(1) Adopting the new photon cross-section data from ENDF/B-VI.

These data were trimmed to cover only the elements from $Z = 1$ to 100 with an effective energy range from 10 KeV to 20 MeV.

(2) Using the improved detector response functions.

The new detector response functions [5] were generated from a modified Monte Carlo simulation code that accounts for (1) the inherent nonlinearity of NaI detectors [2] resulting from variable scintillation efficiencies of different deposited electron energies and (2) the variable flat continuum due to the loss of electrons that escape from the detector surface without depositing all of their energy. These things are accomplished by using an experimental relationship between scintillation efficiency and deposited electron energy and a pseudo electron range, respectively.

(3) Adding the neutron activation backgrounds.

Three background library spectra from neutron activation of the NaI detector crystal, including the I and Na prompt gamma-ray spectra, the ^{24}Na decay spectrum and the ^{128}I decay spectrum for a variety of NaI detectors were measured experimentally by Gardner et al. [4]. Monte Carlo codes have been developed [6] to predict the ^{24}Na and ^{128}I spectra, but the prompt I and Na spectrum has not been successfully modeled as yet. These library spectra have been directly built into the code and can be output as separate neutron activation background library spectra at the end of the simulation in the same form as other library spectra so that they can be applied directly in the PGNAA analysis.

(4) Generating the natural background library spectra.

The three natural background spectra resulting from the K-40, uranium and thorium decay chains are generated separately using the ALI approach with the assumption that these three decay chains are in the equilibrium state in the sample and their impact on the whole sampling process in the simulation is negligible.

(5) Adding the tracking of the annihilation photons from pair production.

In order to make full use of the information contained in the measured spectrum, the pair production annihilation photons are dealt with explicitly in the new code.

(6) Adopting a general geometry package.

The new general geometry package is capable of characterizing complex PGNAA analyzers. It has the same input format as the widely used general purpose Monte Carlo code MCNP, so that the same problem can be easily treated with both codes.

5. Results

The experimental configuration of a prototype on-line coal PGNAA analyzer is schematically shown in Fig. 1. It is composed of a Cf-252 radioisotope neutron source, a $6'' \times 6''$ NaI (Tl) detector, and a rectangular coal chute located between the source and detector. The cylindrical analyzer body is filled with neutron moderation and shielding materials such as polyethylene, paraffin, lead, etc. With this system, the gamma-ray spectra of six coal samples were obtained with the sulfur compositions shown in Table 1. They were originally measured for verifying the MCLLS algorithm by using the previous CEARPGA code to generate the elemental library spectra. The assumed average composition of the six coal samples is given in Table 2.

Comparisons among the spectra obtained from the calculations by the new CEARPGA code that implements the ALI approach, the previous CEARPGA code that used the expected value splitting technique (EVS), the MCNP code and the experiment are given in Fig. 2. The simulation histories were 200,000 for the new CEARGPA

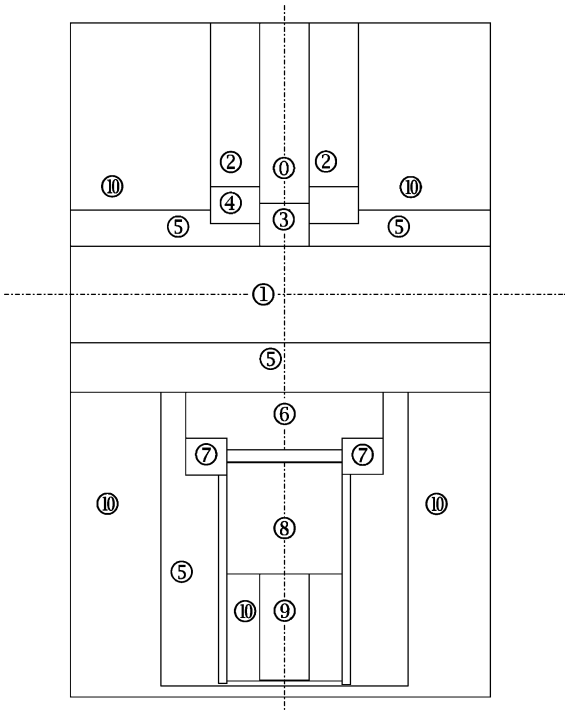

Fig. 1. Schematic of the prototype PGNAA coal analyzer. ① is Coal sample, ② is polyethylene, ③ is bismuth with neutron source, ④ is air, ⑤ is aluminum, ⑥ is Lithium-polyethylene, ⑦ is lead, ⑧ is NaI detector, ⑨ is SiO_2 (PMT) and ⑩ is paraffin.

Table 1
Sulfur content in the coal samples

Coal sample	Sulfur (%)
1	0.57
2	0.68
3	0.36
4	0.78
5	0.61
6	1.19

code, 2 million for the previous CEARPGA code and 20 million for MCNP. Note that the experimental spectrum shown here is only one of six coal sample spectra. From this figure, it is clear that the big weight peaks previously observed around channel 200 in the spectrum calculated from the previous CEARPGA code have been successfully eliminated due to the implementation of the ALI approach. The new calculated spectrum exhibits excellent agreement with the measured spectrum with respect to the corresponding energy peak positions and heights as well as the overall spectral shape. However, an overall spectral upward shift is observed for all simulated spectra including the one obtained from the MCNP code. This may be caused by the following factors: (1) a difference of the coal sample compositions used for the Monte Carlo simulations from that of the experiment, (2) a difference in the geometry description from the actual experiment, (3) a difference of the Monte Carlo and actual structural material compositions, (4) a difference in the assumed and actual source intensity and (5) some as yet unknown error. Comparing the spectrum calculated from the new CEARPGA code to that obtained from the MCNP code, it can be seen that the differences are very small. Especially in the lower range up to channel 450, they agree very well. This indirectly demonstrates that the result calculated from the new CEARPGA code is reasonable and more accurate compared with that obtained from the previous CEARPGA code, which exhibits more shift.

Table 2
The assumed average elemental composition for the six coal samples used for Monte Carlo simulation with a density of 0.86 g/cm^3

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

It needs to be pointed out that the MCNP calculated spectrum was Gaussian spread in the same way as that built into the CEARPGA code. However, the peak shapes in the higher channels appear to have a larger resolution and variance. It turns out that the problem is in the statistics of peak production in the high energy part of the spectra.

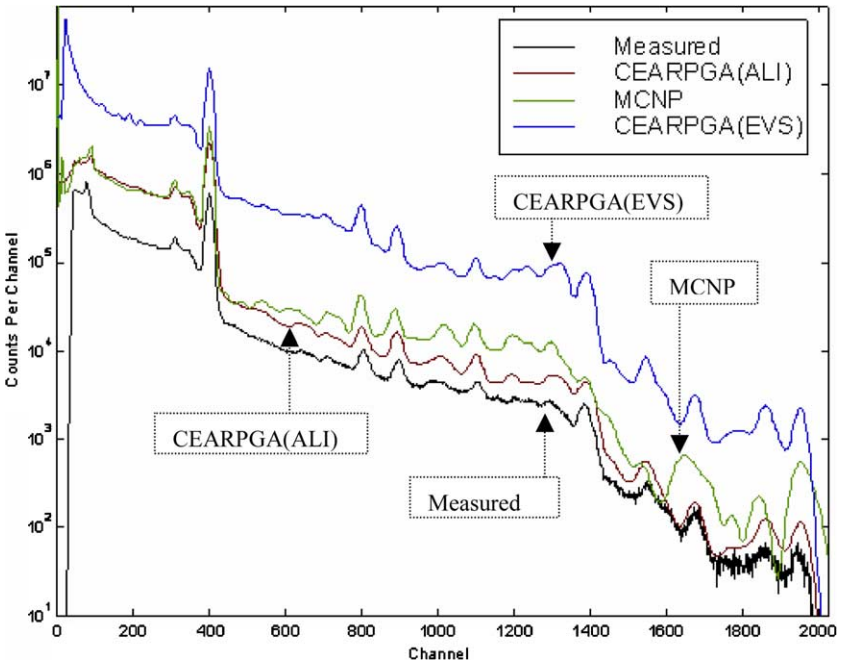

Fig. 2. Comparisons of the calculated coal sample spectra from different Monte Carlo codes and the experimental spectrum.

Since stratified sampling is not being employed in the MCNP code, gamma-ray peaks with very low probability, like the high energy peaks from nitrogen, have very poor variance. This shows up in the very high energy part of the spectra which are three orders of magnitude less intense than the low energy part of the spectrum. In addition, the peak at about channel 1400 which is from the Na and I prompt gamma rays within the NaI detector is not generated by the MCNP code. Therefore, the peak shapes of the CEARPGA code calculated spectra are more like the measured ones. This indicates that the new CEARPGA code is valid in modeling PGNAA analyzers and thus can be used to generate valid elemental library spectra for the MCLLS approach.

The library spectra generated by the new CEARPGA code compared to the previous one generally exhibits higher values in the low energy part of the spectrum. This is consistent with the elimination of the big weight problem in that this is the part of the spectrum where the big weights tend to occur. Two sample library spectra that illustrate this for the two codes are shown for the hydrogen and nitrogen library spectra in Figs. 3 and 4.

6. Discussion and conclusions

The analog linear interpolation approach has been implemented in the new CEARPGA code. In addition, some other improvements to this code have also been introduced, including: (1) adopting the latest photon cross-section data; (2) using the improved detector response function; (3) adding the neutron activation backgrounds; (4) generating and using the individual natural background libraries; (5) adding the tracking of annihilation photons; and (6) adopting a general geometry package.

Comparisons of the simulated result from the new CEARPGA code with those calculated from the previous CEARPGA code, the MCNP code and the experimental data show that the ALI approach has successfully eliminated the big weight problem encountered in the previous CEARPGA code while improving the code efficiency and

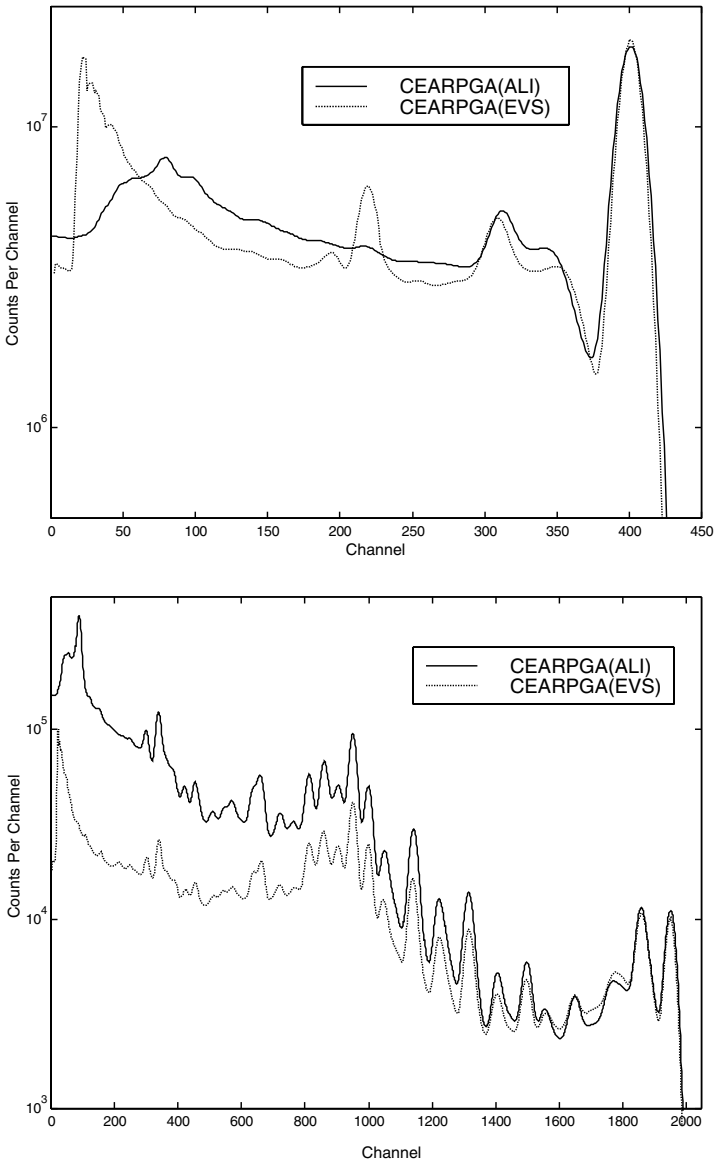

Fig. 3. Hydrogen library spectra for previous (EVS) and present (ALI) CEARPGA codes.

accuracy. Therefore, the new CEARPGA code can be used to generate valid elemental library spectra for PGNAA analysis.

In the future weight windows splitting for the gamma-ray-transport can be added to the CEARPGA code to improve its efficiency even more.

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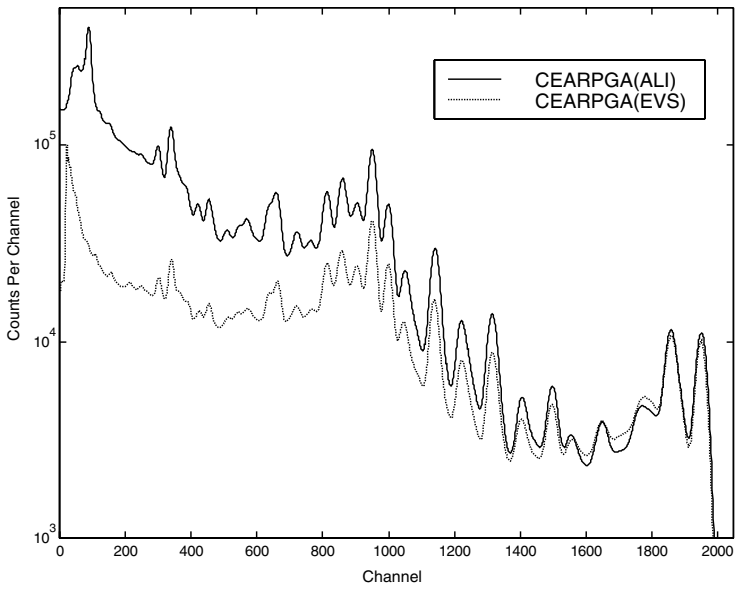

Fig. 4. Nitrogen library spectra for previous (EVS) and present (ALI) CEARPGA codes.

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