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Computer Code Abstract

CEARCPG: A Monte Carlo Simulation Code for Normal and Coincidence Prompt-Gamma-Ray Neutron Activation Analysis

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Abstract—*A new Monte Carlo code named CEARCPG has been developed to generate both the normal and coincidence library spectra for the prompt-gamma-ray neutron activation analysis (PGNAA) inverse analysis problem. A new algorithm for sampling the neutron-induced prompt gamma rays has been developed and implemented within the CEARCPG code, making it possible to calculate the coincidence spectrum by the Monte Carlo method. Compared to the previous code CEARPGA II, which was for normal PGNAA, several improvements have been made, including implementation of the coincidence spectrum simulation and elimination of the "big weight" problem by implementing a new algorithm to generate prompt gamma rays. Several experiments have been carried out to benchmark the new CEARCPG code, and simulation results are also compared with MCNP5 calculations.*

I. INTRODUCTION

Prompt-gamma-ray neutron activation analysis (PGNAA) is a nondestructive, rapid analytical technique that can be used on bulk material streams. It uses the prompt gamma rays emitted during neutron irradiation. Usually, the prompt-gamma-ray spectrum has a low signal-to-noise ratio (SNR) due primarily to a very high background. The background interferences are from the gamma rays emitted by the neutron source; the prompt gamma rays from the structural materials of the PGNAA device; the gamma rays which are from the naturally occurring radioisotopes K-40, U, and Th; and the prompt and radioisotope gamma rays (from ^{24}Na and ^{128}I) which are produced from the activation of the NaI detector (if used) by the neutrons from the source. A possible improvement is to change the traditional PGNAA detection technique by introducing a new gamma-gamma coincidence technique.[2,3] The prompt gamma rays from all elements are emitted in coincidence except that from hydrogen. Research on the coincidence measurement started at the Center for Engineering Applications of Radioisotopes (CEAR) several years ago. Several feasibility studies have already been done and have shown that this approach can effectively increase the SNR by significantly depressing the background interferences.

Before this work, two specific-purpose Monte Carlo codes were developed at CEAR. They were the CEARPGA I and CEARPGA II codes.[4] Both of these codes were developed for the Monte Carlo–Library Least-Squares[5–7] (MCLLS) inverse analysis approach, which has been proposed at CEAR for the elemental analysis of bulk samples. Several variance reduction techniques were applied in these two codes such as stratified sampling, correlated sampling, and the detector response function approach. The analog linear interpolation (ALI) approach was used in the CEARPGA II code in order to address the "big weight" problem, which was a problem in the CEARPGA I code caused by using the expected value splitting technique. However, both of those codes are designed to simulate the normal PGNAA response. The algorithm used in those two codes limits the possibility for those two codes to be developed for coincidence gamma-ray spectrum simulation. Other general-purpose Monte Carlo codes, such as MCNP, cannot be used for coincidence gamma-ray spectra simulation since they use the same algorithm as that used in the CEARPGA I and CEARPGA II codes. Normally, the neutron-induced prompt gamma rays are sampled from a

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precalculated prompt-gamma-ray table that contains individual prompt-gamma-ray energies and their intensities. Statistically, the sampling of individual prompt gamma rays is essentially correct for normal, single gamma-ray spectrum simulation. Only the capability for simulating detector sum counts is lost with this approach. The drawback of this gamma-ray selection algorithm is that it loses the coincident relationship among the prompt gamma rays. Actually, this information is the key ingredient for coincidence measurement, and it represents the true physical phenomenon. In order to solve this problem, a new algorithm had to be developed.

In this work, such a new algorithm is developed and used for sampling the coincident prompt gamma rays, and a new specific-purpose Monte Carlo code named CEARCPG has been developed based on this new algorithm. This coincidence PGNAA approach has been carefully studied using the Monte Carlo code CEARCPG and this method, and all the simulated results have been benchmarked with experiments.

II. THE MONTE CARLO CEARCPG CODE

II.A. Features of the CEARCPG Code

The CEARCPG code is a specific-purpose, continuous-energy, generalized-geometry Monte Carlo code that can be used to simultaneously simulate both the normal (single) and the coincidence spectra for PGNAA. The user needs to create the input file that will be read by CEARCPG. The information in the input file contains (a) the geometry specification, (b) the description of materials, (c) neutron source information, and (d) information related to the tallies. The output files include the singles library spectra of all the elements in the sample and the coincidence library spectra if the coincidence measurement is called for in the input file. These calculated library spectra are used in the input file for the MCLLS fitting.

The main features of the CEARCPG code are as follow:

1. The neutron library includes 97 isotopes of practical interest. The neutron energy ranges from 10^{-11} to 20 MeV.

2. Three main neutron libraries are used for neutron transport:

   a. the neutron cross-section libraries, which are extracted from the ENDF/B-VI.8 300K and the JENDL-3.3 300K libraries

   b. the (n,γ) scheme libraries, which are used to sample the prompt gamma rays from the neutron capture reaction

   c. the neutron inelastic scattering libraries, which are used to sample the gamma rays that are produced through the neutron inelastic scattering reaction.

3. The library for gamma rays includes the elements $Z = 1$ to 100, which come from the EPDL97 library. The gamma-ray energy range is from 0.01 to 20 MeV.

4. Most of the CEARCPG code input cards take the same form as those used in MCNP (Ref. 8). By using the platform VisED of MCNP5, it is easy for the user to design and check the geometry used in the simulation.

The formats of the neutron cross-section data and nuclide structure data are the same as the ENDF and ENSDF nuclear data format. In MCNP, the nuclear data must be processed into the desired format using the NJOY code. It is easy for users to update. For CEARCPG code usage, any new nuclear data are copied directly without having to conform to a specific data format.

The CEARCPG code inherited several variance reduction techniques from the CEARPGA I and CEARPGA II codes. Some new techniques were also added. They are the following:

1. *The stratified sampling technique*—this forces neutron interactions with all the elements. It guarantees that all the calculated elemental library spectra have the same statistical significance regardless of their amount in the sample.

2. *The detector response function*[9]—This is used to convert the gamma-ray flux incident on the detector surface into pulse-height spectra. This technique is considered to be very powerful and can save as much as 50% of the total tracking time of each neutron history. The detector response functions are usually more accurate than tracking within the detector since the Monte Carlo simulation code used to calculate the detector response function has more accurate models.

3. *Truncated probability density function (pdf)*—This approach forces the neutron to interact before it escapes from the system boundary or from the boundary of a sample cell. This option is controlled by the input deck.

4. *Particle splitting*—This is used to increase the interaction rate when the neutron interacts with the elements. This option is controlled by the input deck.

5. *Implicit capture*—Using this technique ensures that the neutron will never be killed by neutron absorption. The neutron weight must be adjusted for this technique based on the absorption and total cross sections.

II.B. Algorithm Used to Sample Neutron-Induced Prompt Gamma Rays

In PGNAA, photons are generated from neutron inelastic or neutron capture reactions. For most of the

existing Monte Carlo codes, such as MCNP, photons are sampled from precalculated photon abundance tables. This approach is essentially correct for the problem of single gamma-ray pulse-height spectrum simulation. However, the coincidence relationship between these neutron-induced photons will be lost by using this approach, and true summing events are not accounted for in this case. A new algorithm has been developed in the CEARCPG code to generate the neutron-induced photons that are emitted in coincidence. The primary nuclear data used to sample these neutron-induced photons are from the ENSDF data libraries. Figure 1 shows the typical nuclei structure of ^{13}C that is produced from the $^{12}C(n,\gamma)$ reaction. From that scheme, the following information is supplied for prompt gamma-ray sampling: (a) the energy and intensity of each prompt gamma ray, (b) the energy of each excited level, (c) the lifetime of each excited level, and (d) the structure information for each prompt gamma ray. For the neutron capture reaction, the highest energy level equals the Q value of the reaction. The nuclei will deexcite from the highest level until it reaches the ground level through various decay cascades. The most common way of decaying is to either emit photons or to undergo internal conversion.

Since there are no photons emitted in internal conversion, the CEARCPG code considers only the case of photon emission. If there is some internal conversion from a given level, the photon weight needs to be adjusted. Since the lifetime of each level is extremely short, all the photons can be regarded as being emitted simultaneously. At each energy level, the emitted photons are sampled based on their relative intensity.

Sampling the photons from the neutron inelastic scattering reaction is a little more complicated than sampling from the neutron capture reaction since the photons emitted from neutron inelastic scattering depend on the excited level. The cross-section data for each excited level are used first to determine which level will be sampled. These cross-section data are from the ENDF nuclear data library. When the excited level is determined, the nuclei structure data for the neutron inelastic scattering reaction are used to sample the appropriate gamma rays.

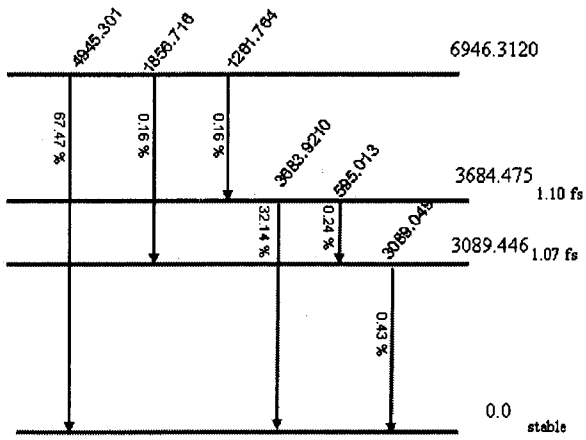


Fig. 1. Nuclei structure of ^{13}C from the neutron capture reaction.

Compared to the algorithm used in the ALI scheme used in the CEARPGA II code, this new algorithm is more accurate since there is no error introduced by interpolation. It is also faster in that only true gamma rays are tracked. No further interpolation is needed, and this new algorithm makes it possible to simulate the coincidence spectrum.

II.C. Simulation of NaI Detector Neutron Activation

NaI detector activation produces the prompt gamma rays from both Na and I and the decay gamma rays from the radioisotopes ^{128}I and ^{24}Na. These NaI detector activation spectra are calculated based on this new algorithm. Compared to the calculation results of the CEARPGA II code, the new simulated spectra are closer to experimental spectra, partly because the necessary data are now more complete.

II.D. Neutron Source

The default neutron source in the CEARCPG code is the ^{252}Cf spontaneous fission source. CEARCPG also supports any user-defined neutron source. Since the ^{252}Cf spontaneous fission source emits not only fission neutrons but also fission gamma rays, both of these are taken into account. The energy distribution spectrum of fission neutrons is assumed to be a Watt fission spectrum. The energy distribution spectrum of fission gamma rays is from the experimental work by Verbinski.[10] Both neutrons and photons are emitted isotropically. Since there is more than one fission gamma ray emitted per spontaneous fission, the multiplicity[11] of fission gamma rays is also taken into account. These fission gamma rays might contribute to the coincidence spectrum.

II.E. Neutron Transport

The CEARCPG code considers only neutron elastic scattering, neutron inelastic scattering, and the neutron capture reaction, which are the major reactions for nonfissionable material. The neutron weight is adjusted based on the cross sections at each collision point. The Russian roulette game is played to terminate the small weight neutrons. The analog method is used to sample between the neutron elastic and inelastic scattering reaction. Compared to the CEARPGA I and CEARPGA II codes, all the neutron cross-section data are from ENDF/B-VI.8 300K and JENDL-3.3 300K with their original format. The angular distribution of scattered neutrons is also considered based on the ENDF nuclear data library.

Typically, the distribution data are given in the center-of-mass system for neutron elastic and inelastic scattering reactions and are dependent on incident energy. The sampled cosine of the angle between incident and exiting particle direction is needed to transfer from the center-of-mass system to the laboratory system.

Since most prompt gamma rays are emitted from the neutron capture reaction, the variance reduction technique of implicit capture is used to increase the number of prompt gamma rays. In the real physical case, a neutron history would be terminated if it was absorbed by the nucleus. With implicit capture, the neutron is never killed by the neutron capture reaction. The neutron weight must be adjusted based on the pertinent cross sections. Normally, the neutron will be killed only by Russian roulette based on the neutron weight or if it escapes from the system when the truncated pdf technique is not selected for use.

Photons generated from neutron collisions inherit the weight of the neutron that produces it. That created photon weight is

$$W_p = \frac{W_n \sigma_\gamma (1 - \alpha_k)}{\sigma_T} , \quad (1)$$

where

W_p = photon weight

W_n = neutron weight

σ_γ = photon production cross section

σ_T = total neutron cross section

α_k = internal conversion fraction of the excited level k.

The ENSDF nuclear data library includes α_k for some isotopes. If there is no internal conversion factor in the ENSDF data library, it is calculated by an appropriate balance.

The stratified sampling technique is always used when sampling the prompt gamma rays from the elements. In analog Monte Carlo simulation, only one element will be sampled per neutron collision. In stratified sampling, every element in the sample will be forced to interact with the incident neutron. This technique assures that every calculated elementary library spectrum has the same statistical significance independent of its concentration in the sample. It also represents a very efficient use of the neutron histories. For the gamma rays from neutron inelastic scattering, the analog method is used.

II.F. Photon Transport

The photon cross-section data were extracted from the EPDL 97 data library and were processed into three separate tables. They 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. At present, the CEARCPG code takes into account only photoelectric effect, Compton scattering, and pair production interactions. There is no X-ray or Bremsstrahlung radiation generation and no Rayleigh scattering. The CEARCPG code models the transport of all the gamma rays from

1. the neutron capture reaction
2. the neutron inelastic-scatter reaction
3. the neutron source fission reaction (a background source)
4. other backgrounds, including the three from natural radioisotopes, the three from NaI neutron activations, and photons from the structural materials.

The photon is terminated if it escapes from the system or is killed by Russian roulette. If the photon reaches the detector, it will be tallied (it is weighted by the total efficiency calculated from the total linear attenuation over its specific path through the detector) and convoluted with the detector response function to convert the incident photon flux tally into a pulse-height spectrum tally.

Table I compares the MCNP5 and CEARCPG codes.

III. BENCHMARK EXPERIMENTS

Three experiments were designed to benchmark the normal (single) and the coincidence spectra simulation of the CEARCPG code. These are the Energy Technologies, Inc. (ETI) prototype experiments, which benchmark the normal case, and the sulfur and mercury experiments, which benchmark the coincidence case.

III.A. Energy Technologies, Inc., Prototype Experiments

The spectra from six coal samples provided by ETI were obtained from a PGNAA coal analyzer prototype. The sulfur amounts in these six coal samples are presented in Table II. Each coal sample spectrum was accumulated for 5 h with a 2.9-μg ^{252}Cf spontaneous fission neutron source. The Monte Carlo model for the coal analyzer geometry is schematically shown in Fig. 2, which consists of a ^{252}Cf spontaneous fission neutron source, a 15.24- × 15.24-cm (6- × 6-in.) 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, and lead. In Monte Carlo simulations, the average compositions of the six coal samples were used and are shown in Table III. The number of simulation histories used was 2×10^5.

Since there was only one detector used to record the prompt-gamma-ray spectra in the prototype, only single

TABLE I

Comparison Between the CEARCPG and MCNP Codes

	CEARCPG	MCNP
Nuclear data	ENDF/B-VI ENSDF EPDL with original format	ENDF/B-VI ENSDF with NJOY format EPDL
Neutron interaction	Neutron capture reaction Neutron elastic scattering reaction (free gas thermal treatment) Neutron inelastic scattering reaction $(n, n'\gamma)$	Same All inelastic scattering reaction, such as (n, n') $(n,2n)$, etc.
Generation of neutron-induced photons	Sampling from isotope scheme	The number is function of neutron weight, photon limit weight, photon production cross section, etc.
Photon interaction	Simple physics treatment	Simple physics treatment and detailed physics treatment
Variance reduction technique	Stratified sampling, DRF, etc.	General

TABLE II

Sulfur Experimental and MCLLS Calculated Amounts in Six Coal Samples

Coal	Sulfur Amount [Weight Fraction (%)]	
	Experimentally Measured	MCLLS Calculated
1	0.57	0.59 ± 0.03
2	0.68	0.745 ± 0.01
3	0.36	0.492 ± 0.02
4	0.78	0.707 ± 0.03
5	0.61	0.543 ± 0.02
6	1.13	1.06 ± 0.01

TABLE III

Elemental Composition for Coal Samples Used for Monte Carlo Simulation

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

spectra are simulated in this case. The general-purpose Monte Carlo code MCNP5 was also used to simulate the coal sample with the same composition as that in Table III. Since the ^{252}Cf spontaneous fission source emits not only fission neutrons but also fission associated gamma rays, two independent cases, one for neutrons and one for

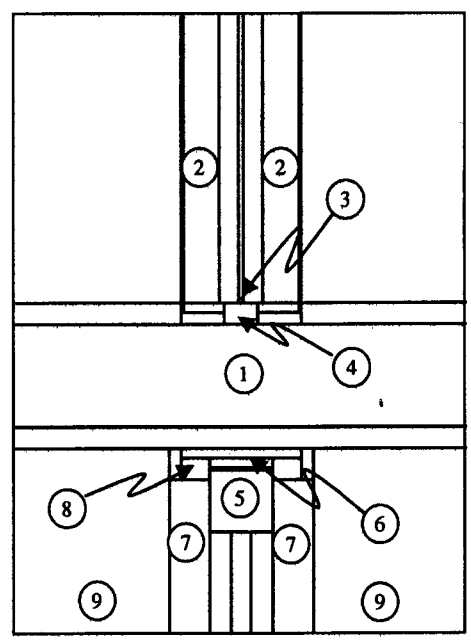


Fig. 2. Schematic diagram of the ETI prototype coal analyzer: 1 = coal sample, 2 = polyethylene, 3 = ^{252}Cf fission neutron source, 4 = bismuth, 5 = NaI detector, 6 = lithium-polyethylene, 7 = aluminum, 8 = lead, and 9 = paraffin.

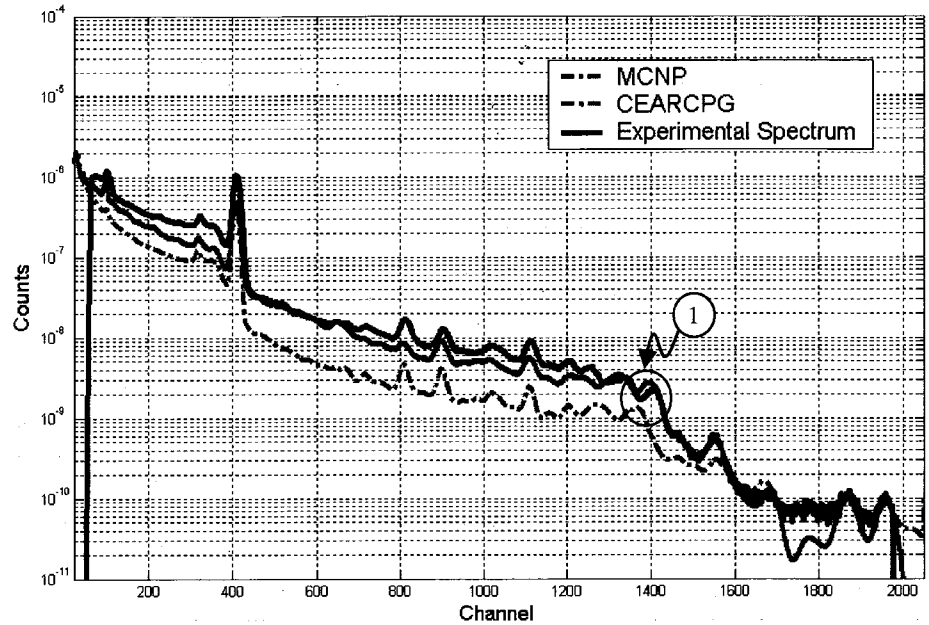


Fig. 3. The singles spectrum of the ETI prototype for a coal sample: 1 indicates the peak from NaI detector activation.

gamma rays, were simulated with different particle sources to describe the total response. The sum of these two simulated spectra was taken to give the final spectrum that is plotted in Fig. 3. All three spectra are normalized at the nitrogen peak. (Note that it should not be expected that the simulated and experimental spectra should match exactly since most of the background spectra are not accounted for in this case and only the average composition of each coal sample is used.) All the simulated spectra are normalized at the nitrogen peak. It is clear that the MCNP code does not predict the prompt-gamma-ray peak that was neutron activated within the NaI detector, which is labeled 1 in Fig. 3. The simulated elemental library spectra serve as the input files to the MCLLS fitting to calculate the composition of sulfur in the samples. The fitting reduced chi-square values for the six coal samples ranged from 13 to 69, which are a factor of ~2 better than those reported by Zhang and Gardner.[4] The calculated sulfur results are presented in Table II. The reduced chi-square value for the six sulfur amounts was 19, compared to 45 for the simulations by Zhang and Gardner,[4]

III.B. Pure Sulfur Sample Experiments

The arrangement for this experiment is schematically plotted in Fig. 4. Two 15.24- × 15.24-cm (6- × 6-in.) NaI detectors were used to record the single and coincidence spectra. A spontaneous fission neutron source of ^{252}Cf was used. Wax is used to slow down the fission neutrons to thermalize them before they interact with the sulfur. The experimental spectrum is plotted in Fig. 5.

For comparison purposes, two calculated spectra are also plotted in Fig. 5. One is calculated by using the MCNP code, and the other is calculated with the CEARCPG code. Both calculated spectra are normalized with each other at the hydrogen peak, but not with the experimental spectrum. The number of neutron histories used for the MCNP simulation was 1×10^8, while that for the CEARCPG code was 1×10^6. Since neither of the calculated spectra are normalized to the experimental spectra, this leads to some of the observed discrepancy between the experimental and simulated spectra. The primary discrepancy between the simulated and

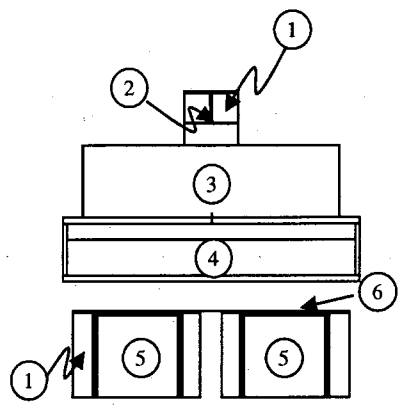


Fig. 4. Schematic diagram for pure sulfur sample experiment: 1 = lead, 2 = ^{252}Cf spontaneous fission source, 3 = wax, 4 = sulfur sample, 5 = NaI detector, and 6 = lithium-polyethylene.

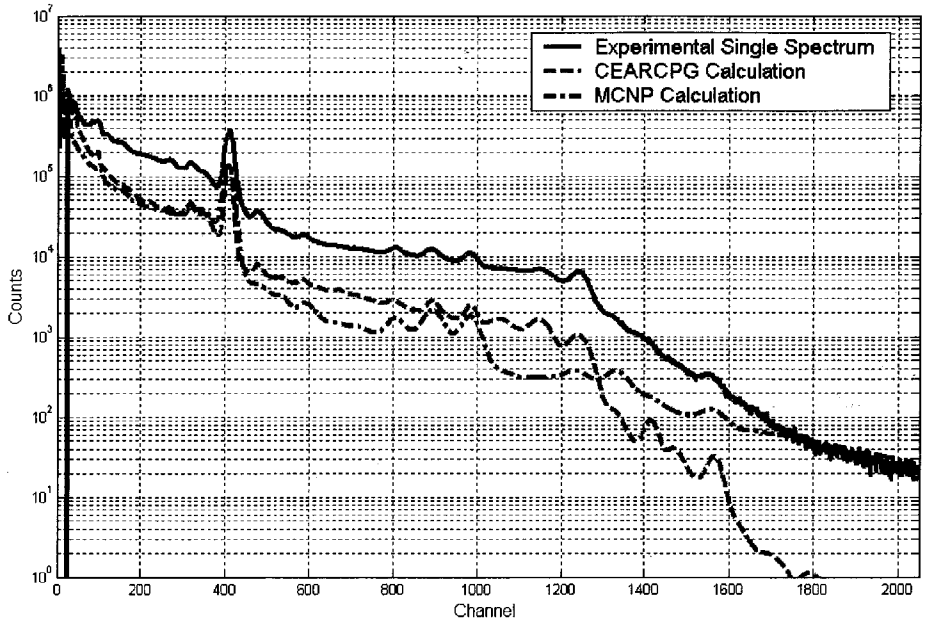


Fig. 5. The experimental and simulated singles spectra of the pure sulfur sample.

experimental spectra is caused by the omission of the background in the simulated spectra. This includes the three natural background sources of U, Th, and ^{40}K and the three neutron-activated detector background sources of ^{24}Na, ^{128}I, and the prompt gamma rays of Na and I. These sources of background represent as much as 70 to 80% of the total experimental spectrum.

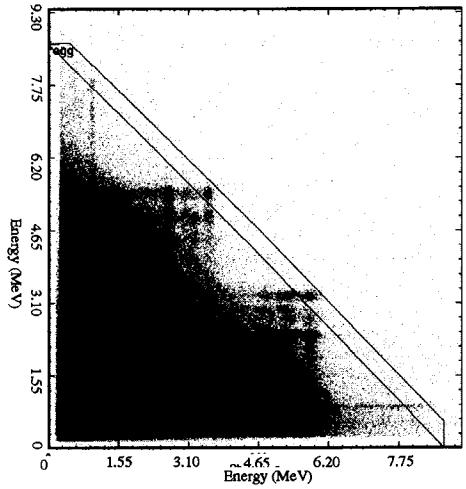
With the SPARROW multiparameter acquisition system,[12] one can obtain two-dimensional coincidence spectra. Such a spectrum is plotted in Fig. 6a, where the coincidence intensity is proportional to the degree of darkness in the plots.

With the two-dimensional coincidence spectrum, a diagonal window area[13,14] can be defined that has the property that pairs of peaks in that window sum to the energy that corresponds to the Q value of the $^{32}S(n,\gamma)^{33}S$ reaction (8.641 MeV). According to the nuclei scheme[15] of ^{33}S, the spectrum delineated by the diagonal Q-value window contains the pairs of gamma rays whose energies add up to the ^{33}S Q value. For the ^{33}S reaction case, there are two pairs of these energies, at 7.800 and 0.841 MeV and at 5.420 and 3.221 MeV. Therefore, one can expect these four peaks in the spectrum of the Q-value projection. The peak resolution (defined as the full-width at half-maximum) for the 5.420-MeV peak in the normal spectrum was 3.5%, while in the Q-value diagonal projection spectrum, this resolution improves to 2.48%. In addition to the lower background, the peak resolution is also improved by using this method. This is consistent with the original findings of Hoogenboom.[13]

The calculated two-dimensional coincidence spectrum is plotted in Fig. 6b. The Q-value projection approach was also applied to this spectrum. The projection spectra are plotted in Fig. 7 for comparison. The two calculated spectra were fitted by least squares, and the reduced chi-square value was found to be 2.3.

III.C. Pure Mercury Sample Experiments

This experiment was carried out on the PULSTAR research reactor at North Carolina State University. The neutron flux was $\sim 1.8 \times 10^6$ n/s·cm^{-2}. The geometry is plotted in Fig. 8. Natural mercury has seven stable isotopes. Considering the neutron capture cross sections and the isotope abundances, almost 94% of the prompt gamma rays of mercury are from ^{199}Hg. The general-purpose Monte Carlo code MCNP was also used for comparison purposes. Two mercury nuclear data sets in the MCNP data library were tested. They are the 80000.40c and 80000.42c data sets. The calculation results show that the mercury nuclear data set used in the MCNP code does not seem right. Therefore, only the experimental and calculated spectra from the CEARCPG code are shown in Fig. 9. The experimental two-dimensional coincidence spectrum and the calculated two-dimensional spectrum are plotted in Fig. 10, and the Q-value diagonal projection approach was applied. The energy of the diagonal window equals the Q value of the $^{199}Hg(n,\gamma)^{200}Hg$ reaction, which is 8.028 MeV. The projection spectra are plotted in Fig. 11. Three pairs of peaks are observed in the projection spectrum. According to the ENSDF nuclear data library, they are at 1.571 and 6.457, 2.639 and 5.387, and 3.185 and 4.841 MeV. The calculated projection



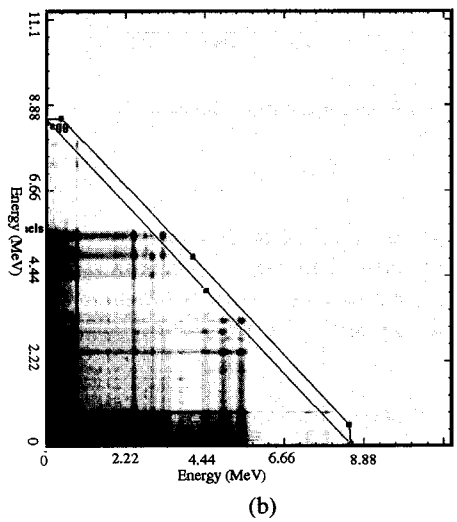


Fig. 6. Two-dimensional spectra of the counts in both NaI detectors for a sulfur sample. (a) The experimental two-dimensional spectrum for a pure sulfur sample. (b) The simulated two-dimensional spectrum for a pure sulfur sample.

spectrum fitted by using a least squares approach gives a reduced chi-square value of 4.3.

III.D. The Monte Carlo–Library Least-Squares Approach Results for the Noncoincidence Case

Figure 12 compares the measured and MCLLS fitted coal spectra. Compared to that obtained with the CEARPGA II code,[4] the result using the CEARCPG code exhibits a better fit in the low-energy region since the CEARCPG code takes into account all the prompt gamma rays regardless of their intensities. All of the six measured coal sample spectra were fitted using the MCLLS algorithm with the CEARCPG code. The reduced chi-

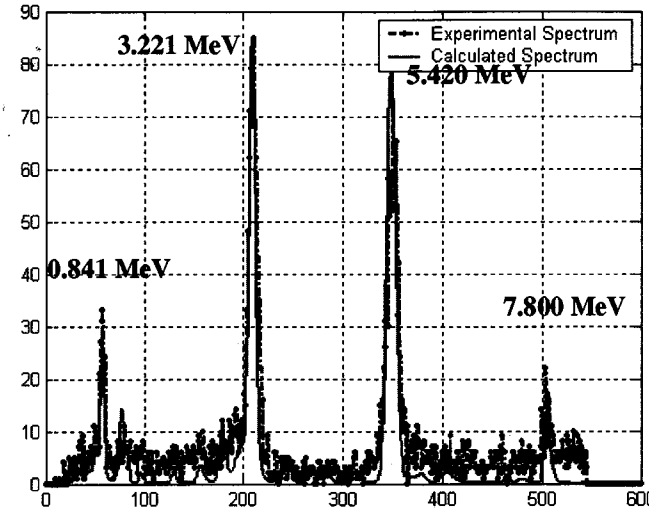


Fig. 7. A comparison of the experimental and calculated spectra for sulfur.

squares obtained are from 15 to 60. According to the residuals spectrum, which is plotted in Fig. 13, the maximum residual peak is around the hydrogen peak. We believe this might be caused by a slightly incorrect resolution of the detector response function (DRF) at that energy and/or a slight spectrum gain shift.

IV. DISCUSSION AND CONCLUSION

The CEARCPG code is the first specific-purpose Monte Carlo code that can be used to calculate the coincidence spectra for PGNAA analysis. The algorithm used in MCNP5 and MCPNX (Ref. 16) of sampling individual prompt gamma rays limits their capabilities for simulating coincidence spectra. MCNPX allows the calculation of neutron coincidence from a neutron source

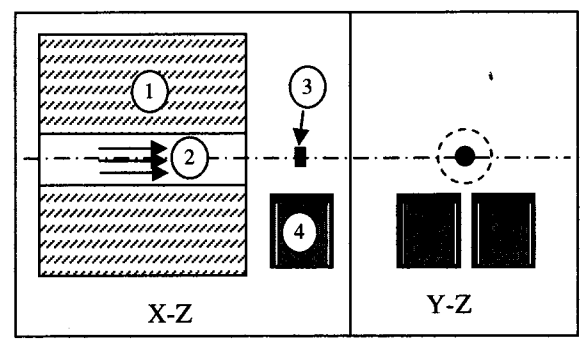


Fig. 8. Schematic diagram for the pure mercury sample experiment: 1 = concrete, 2 = thermal neutron beam, 3 = mercury sample, and 4 = NaI detector.

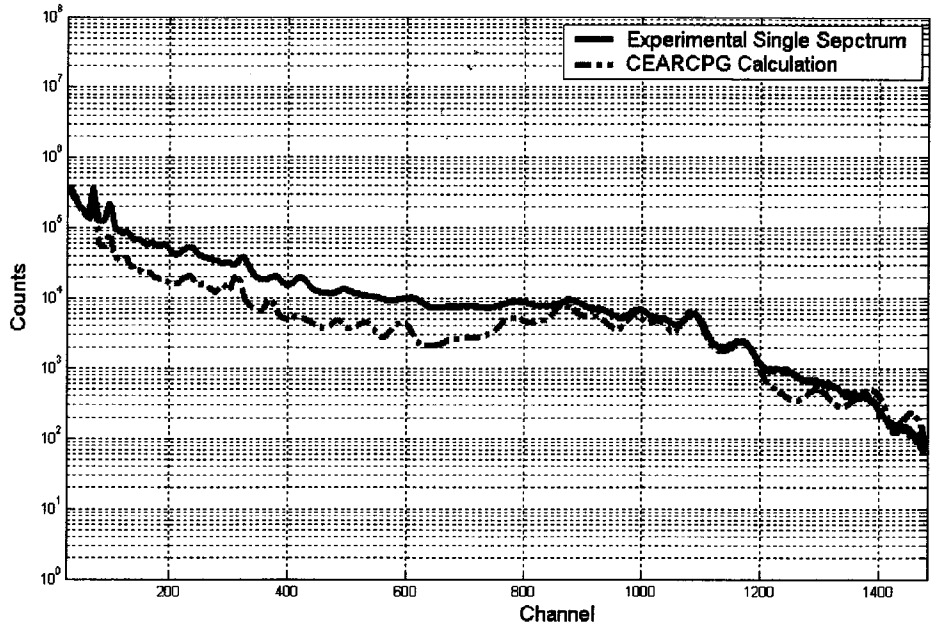


Fig. 9. Comparison of experimental and calculated single spectra for a pure mercury sample.

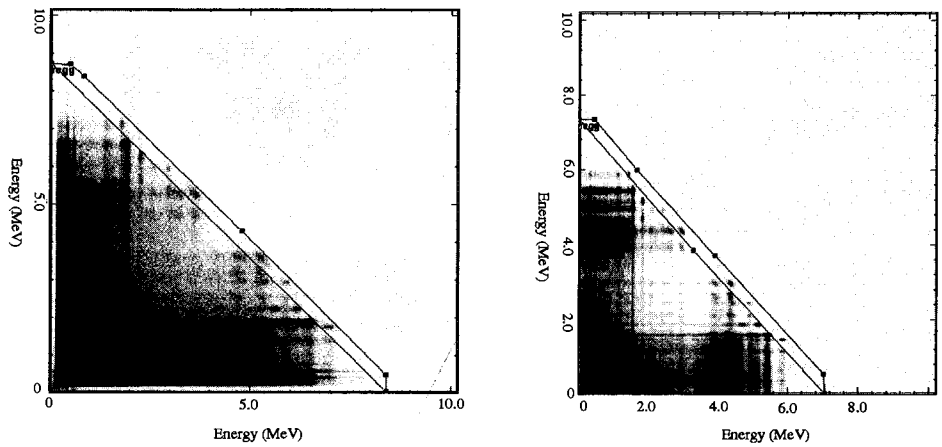


Fig. 10. The two-dimensional coincidence spectra of a pure mercury sample. (a) The experimental two-dimensional spectrum for a pure mercury sample. (b) The simulated two-dimensional spectrum for a pure mercury sample.

that emits multiple neutrons simultaneously. It also allows a user to calculate a coincidence spectrum for single gamma rays, each of which deposits its partial energy in several detectors. For coincidence PGNAA analysis, the true coincidence event is not from a single gamma ray but from multiple prompt gamma rays emitted in coincidence from the neutron capture reaction. Theoretically, since the algorithm used to sample prompt gamma rays in the MCNPX code is the same as that used in the MCNP code, this also limits the MCNPX code in simulating the coincidence spectrum for coincidence PGNAA analysis.

The CEARCPG code was developed at CEAR and has been benchmarked by several experiments. The benchmark experiments show that the simulation results are accurate. These experiments also indicate that the coincidence PGNAA analysis could significantly reduce the interference from natural backgrounds, the neutron-activated NaI backgrounds, structural materials, the gamma rays from the neutron source, and the hydrogen peak and could significantly improve the SNR.

The CEARCPG code can also be used for coincidence PGNAA application design. For example, it can be used to optimize the coincidence PGNAA application.

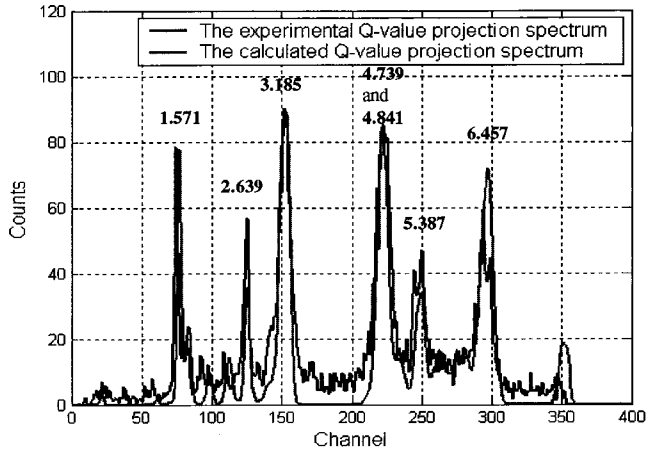


Fig. 11. The Q-value diagonal projection spectrum for a pure mercury sample.

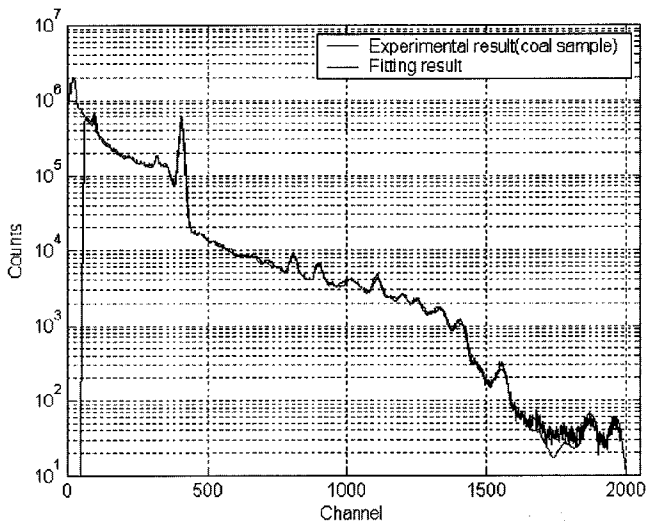


Fig. 12. The experimental versus the MCLLS fitted coal spectrum.

It is well known that the biggest disadvantage of the coincidence technique is that it will significantly lower the signal counting rate as well as that of the background. To combat this problem a new coincidence PGNAA application arrangement has been proposed and optimized using the CEARCPG code. The new coincidence PGNAA arrangement is shown schematically in Fig. 14. In this geometry, one 15.24- × 15.24-cm (6- × 6-in.) NaI detector is surrounded by six 7.62- × 7.62-cm (3- × 3-in.) NaI detectors, which work as "trigger" detectors. The six small NaI detectors are a connected assembly. If there is energy output from any one of the small detectors and energy output from the big detector simultaneously, one coincidence count will be recorded. Compared to the old geometry arrangement in which

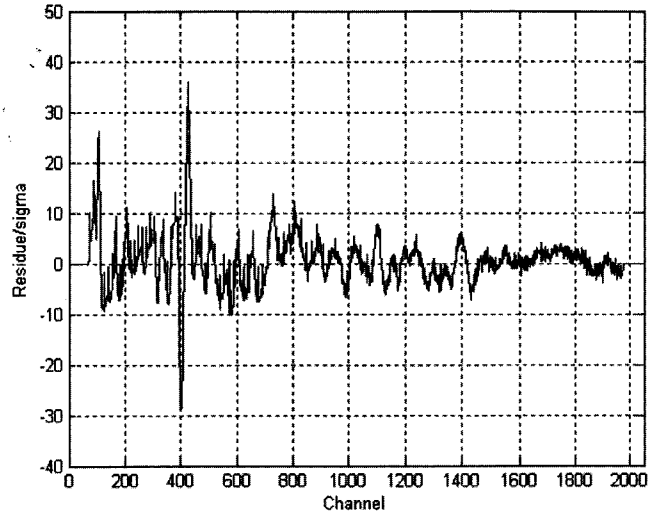


Fig. 13. The residuals between the experimental and MCLLS fitted spectra.

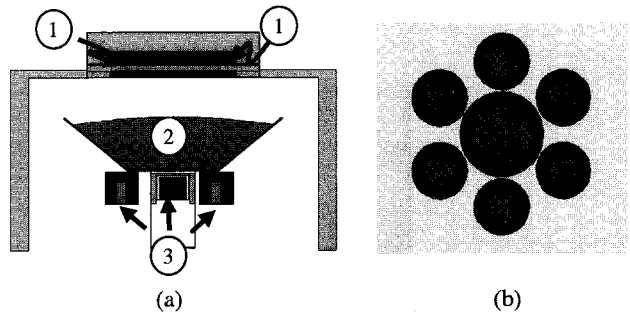


Fig. 14. The optimized coincidence PGNAA application: 1 = neutron source, 2 = coal on the conveyor belt, and 3 = NaI detector array. (a) X-Z plane; (b) X-Y plane.

two 15.24- × 15.24-cm (6- × 6-in.) NaI detectors are placed side by side, the coincidence efficiency of this new PGNAA coincidence arrangement is 3.17 times higher.

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REFERENCES

1. R. P. GARDNER, E. S. EL-SAYYED, Y. ZHENG, S. HAYDEN, and C. W. MAYO, "NaI Detector Neutron Activation

Spectra for PGNAA Application," *Appl. Radiat. Isotopes*, **53**, 483 (2000).

2. R. P. GARDNER, C. W. MAYO, E. S. EL-SAYYED, W. A. METWALLY, Y. ZHENG, and M. POEZART, "A Feasibility Study of a Coincidence Counting Approach for PGNAA Applications," *Appl. Radia. Isotopes*, **53**, 515 (2000).

3. W. A. METWALLY, R. P. GARDNER, and C. W. MAYO, "Elemental PGNAA Analysis Using Gamma-Gamma Coincidence Counting with the Library Least-Squares Approach," *Nucl. Instrum. Methods Phys. Res. B*, **213**, 394 (2004).

4. W. ZHANG and R. P. GARDNER, "CEARPGA II: A Monte Carlo Simulation Code for Prompt-Gamma-Ray Neutron Activation Analysis," *Nucl. Sci. Eng.*, **151**, 361 (2005).

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

6. 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.*, **2**, 241 (1993).

7. R. P. GARDNER, "Software Design for a PGNAA Coal Analyzer Prototype," Report to Energy Technologies, Inc., North Carolina State University (2000).

8. "MCNP—A General Monte Carlo N-Particle Transport Code Manual," Version 5, Vol. 1, X-5 Monte Carlo Team, Los Alamos National Laboratory (2003).

9. A. SOOD and R. P. GARDNER, "A New Monte Carlo Assisted Approach to Detector Response Functions," *Nucl. Instrum. Methods Phys. Res. B*, **213**, 100 (2004).

10. V. V. VERBINSKI, H. WEBER, and R. E. SUND, "Prompt Gamma Rays from $^{236}U(n,f)$, $^{239}Pu(n,f)$ and Spontaneous Fission of ^{252}Cf," *Phy. Rev. C*, **7**, 3 (1973).

11. T. E. VALENTINE, "Evaluation of Prompt Fission Gamma Rays for Use in Simulation Nuclear Safeguard Measurement," *Ann. Nucl. Energy*, **28**, 191 (2001).

12. Sparrow Multi-Parameter Data Acquisition System, Sparrow Corporation, http://www.sparrowcorp.com.

13. A. M. HOOGENBOOM, "A New Method In Gamma-Ray Spectroscopy: A Two Crystal Scintillation Spectrometer with Improved Resolution," *Nucl. Instrum.*, **3**, 57 (1958).

14. W. A. METWALLY, C. W. MAYO, X. HAN, and R. P. GARDNER, "Coincidence Counting for PGNAA Application: Is It the Optimum Method?" *J. Radioanal. Nucl. Chem.*, **265**, 2, 309 (2005).

15. Evaluated Nuclear Structure Data File (ENSDF); available on the Internet at http://www.nndc.bnl.gov/ensdf/.

16. "MCNPX Extensions, Version 2.5.0," LA-UR-05-2675, MCNPX Team Group, Los Alamos National Laboratory (2005).