The Monte Carlo Method: Versatility Unbounded In A Dynamic Computing World
Chattanooga, Tennessee, April 17–21, 2005, on CD-ROM, American Nuclear Society, LaGrange Park, IL (2005)

DEVELOPMENT OF A MONTE CARLO CODE FOR COINCIDENCE PROMPT GAMMA NEUTRON ACTIVATION ANALYSIS

Xiaogang Han, Robin. P. Gardner*, Charles W. Mayo, and Weijun Guo
Center for Engineering Application of Radioisotopes (CEAR)
North Carolina State University, Raleigh, NC 27695-7909, USA
hxiaoga@unity.ncsu.edu; Gardner@ncsu.edu;mayocwm@aol.com;wguo@ncsu.edu

ABSTRACT

Prompt Gamma-Ray Neutron Activation Analysis (PGNAA) offers a non-destructive, relatively rapid on-line method for determination of elemental composition of bulk and other samples. However, PGNAA has an inherently large background - that primarily is due to the presence of the neutron excitation source. This includes neutron activation of the detector, gamma rays from the neutron source, and prompt gamma rays from the structure materials of PGNAA devices. This large background limits the sensitivity and accuracy of PGNAA. Since Most of the prompt gamma rays from the same element are emitted in coincidence, the gamma-gamma coincidence technique is being investigated at CEAR to increase detection sensitivity and accuracy in PGNAA by essentially eliminating most of the background. The Monte Carlo code CEARCPG is being developed to predict coincidence counting in coincidence PGNAA and also to optimize the design of coincidence PGNAA gauges that utilize this approach. The CEARCPG Code is a specific-purpose, continuous-energy, generalized-geometry, coupled neutron/photon Monte Carlo transport code. The neutron energy regime is from 10^{-11} to 20 MeV. The photon energy regime is from 0.01 to 20 MeV. The CEARCPG Code can be used to analyze 97 isotopes of practical interest. It is the first Monte Carlo code to properly treat coincidence counting directly that can be used for the investigation of coincidence counting in all PGNAA applications. Benchmark bulk sample experiments have been performed with coal and sulfur samples. Results indicate that the code is accurate and will be very useful in the design and use of coincidence PGNAA devices for all the many applications of this technology.

Key Words: Coincidence PGNAA, Monte Carlo, CEARCPG

1 INTRODUCTION

There are a number of important applications of PGNAA including on-line industrial bulk sample elemental analysis, small sample neutron beam elemental analysis, oil well logging elemental analysis, and aluminum alloy sorting to name a few. CEAR has concentrated on the first of these so far, but is now initiating projects on all of the others. Therefore, the results reported here are primarily on the first application of on-line bulk sample elemental analysis.

The traditional method for measuring the compositions of coal on a conveyor belt involve laboratory analysis of samples weighting on the order of 1g each. Not only is this process slow, tedious and expensive for continuous sampling, but also the heterogeneous nature of coal results in the average compositions of the small sample not being representative of the bulk material. Therefore, a need existed for a measurement method that could determine the amounts of the elements in bulk materials on-line, with large representative samples. The analysis should be performed continuously and automatically on samples that represent significant fractions of mass

*Corresponding author. Tel.: +1-919-515-3387; fax: +1-919-515-5115

flow rates that may exceed several hundred tons per hour. The PGNAA approach was ideal for this application and has been used and continuously improved for some time [1-6]. The PGNAA approach is based on the radiative capture reaction between a neutron and the nucleus of any atom. This reaction is a purely nuclear process, which can be used for the characterization of material elemental composition.

The most effective way of doing this is to observe the gamma radiation emitted by the compound nucleus formed after the neutron capture. Neutron-induced prompt gamma-ray activation analysis (PGNAA) primarily exploits the prompt gamma rays themselves, while neutron activation analysis (NAA) uses the delayed gamma rays from the radioactive daughter nuclei that are produced.

Compared to the traditional chemical analysis, PGNAA is a nondestructive, instantaneous method and applicable to all elements in the sample of interest that have a reasonably large product of the nominal amount present times the radiative capture cross section. However, the spectra of PGNAA applications are relatively complicated. The interferences come from the neutron source, the device structural material (for example hydrogen), the natural background and pulse pile-up summing (distortion) effect. The interferences from the neutron source are listed as follows:

1. Gamma rays emitted by the neutron source(for example: a ^{252}Cf neutron source)
2. Gamma rays produced by neutron interaction from the source with the materials of the detector
3. Gamma rays produced by the non-sample materials of construction around or within the PGNAA analyzer

The background interferences are listed as follows:

1. Potassium-40 which emits the gamma rays with energy of 1.461 MeV
2. The natural uranium and thorium chains which emit gamma rays with energies that range up to 2.614 MeV
3. Cosmic radiation which generates a high-energy continuous distribution of gamma rays.

Since the typical background in PGNAA for bulk samples represents as much as 70 to 85% of the total response, a possible improvement is the use of the γ-γ coincidence technique to remove or minimize these background interferences [7-9]. Fortunately, most of the prompt gamma rays are emitted in coincidence. The coincidence detection of gamma rays (the γ-γ coincidence measurement) is a well established technique in nuclear structure studies [10]. Using the γ-γ coincidence technique for PGNAA analysis is a new idea that is being pursued at CEAR [11, 12].

The present work focuses on the experimental feasibility study of coincidence PGNAA applications. The Monte Carlo method is a powerful tool for investigating the coincidence PGNAA application. It can be used to predict the experimental spectrum and optimize the various coincidence PGNAA applications. The CEARCPG code is being developed at CEAR for this purpose. It is programmed in FORTRAN 90.

2 FEATURES OF THE CEARCPG CODE

The CEARCPG code is a specific purpose Monte Carlo code which can be used for coincidence, anti-coincidence, time-of-flight, and normal (non-coincidence) PGNAA simulation for various applications. The code treats an arbitrary three-dimensional configuration of user defined materials bounded by first and second order surfaces. The main purpose of the CEARCPG code is to generate elemental spectra of bulk samples by using Monte Carlo simulation. These elemental spectra are then used as library spectra to calculate the concentration of each element in the bulk sample by using the Monte Carlo-Library Least-Square (MCLLS) approach that is being developed at CEAR [3, 5, 6]. Besides the normal single spectra for PGNAA application, the CEARCPG code can be also used to generate the total coincidence spectrum as well as the coincidence spectra of each element of interest in the sample. The CEARCPG code includes the following main parts:

1. An initialization part which is used to define the geometry, required neutron cross sections, photon cross sections, angular distributions of scattered neutrons, and nuclei excited structure used to sample the prompt gamma rays.
2. A neutron source part which is used to sample the energy and direction of the source neutrons. The default neutron source is ^{252}Cf. The CEARPGA code supports user-defined neutron sources.
3. A neutron tracking part which is used to track the flight of neutrons and sample collision positions as well neutron interaction types.
4. A prompt gamma-ray sampling part which is used to sample the prompt gamma rays generated by neutron radiative capture reactions and neutron inelastic scattering reactions.
5. A gamma-ray tracking part which is used to track the flight of gamma rays and the collision positions as well as the photon interaction types.
6. A tally part which is used to record the spectra detected by detectors as well as single /coincidence spectra of each element of interest.

The main features of the CEARCPG code are listed as follows:

1. The neutron library includes 97 isotopes of practical interest. The neutron energy ranges from 10^{-11} to 20 MeV. Three sub-libraries are included.
   a) The neutron cross-section library, which is extracted from the ENDF/B-VI. 8 300K and the JENDL-3. 3 300K libraries. The angular distribution data of neutron elastic scattering and neutron inelastic scattering of each isotope is also included in this library,
   b) The (n,γ) scheme library, which is mainly extracted from the ENSDF library. This library is used to sample prompt gamma rays caused by the (n, γ) reaction, and
   c) The neutron inelastic scattering library. This library is used to sample the gamma rays that are induced by neutron inelastic scattering.
2. The library for gamma-rays include the elements Z=1 to 100, which come from the EPDL97 library. The gamma-ray energy range is from 0.01 to 20 MeV.

3. Most of the CEARCPG code input card takes the same form as those used in MCNP. By using the platform VisED of MCNP5, it is easy for user to design and check the geometry part used in the simulation.
4. The following gamma-ray spectra are recorded separately by the CEARCPG code:
   a) Prompt gamma rays from neutron radiative capture interaction in the sample zone
   b) Gamma rays from neutron inelastic scattering interaction in the sample zone
   c) Coincidence gamma rays from the sample zone
   d) Prompt gamma rays from neutron radiative capture interactions in the zones other than the sample region
   e) Prompt gamma rays from neutron inelastic scattering interactions in the zones other than the sample region
   d) Coincidence gamma rays from the zones other than the sample region
   f) Gamma rays from the neutron source
   g) Gamma rays from the natural background due to the ^{40}K decay chain
   h) Gamma rays from the natural background due to the Th decay chain
   i) Gamma rays from the natural background due to the U decay chain
   j) Prompt gamma rays from ^{127}I and ^{23}Na from the NaI detector neutron activation
   k) Decay gamma rays from the ^{24}Na resulting from NaI detector neutron activation
   l) Decay gamma rays from the ^{128}I resulting from NaI detector neutron activation

Cross sections and angular distributions of the scattered neutrons used in the CEARCPG code come from the evaluated nuclear data library ENDF/B-VI and JENDL-3.3. Only neutron elastic scattering reaction, inelastic scattering and neutron radiative capture reactions are accounted for. The evaluated nuclear structure library ENSDF is used to sample the gamma rays generated from the neutron inelastic scattering and neutron radiative capture reactions. The data format used in the CEARCPG code is the same as the format of the ENDF/B-VI and ENSDF library. It is not necessary for the user to transfer the data format as is required when using MCNP. This feature makes the CEARCPG code more flexible and extensible. Thermal neutron scatter is described by the free gas model.

Several variance reduction techniques are used in the CEARCPG code. The main ones are: (1) stratified sampling, which is used to sample the neutron interaction type and all the elements in the sample region; (2) Russian roulette, which is used to terminate the neutron and photon histories based on their weights; (3) the rejection method, which is mostly used to sample the direction of scattered neutrons since most of the angular distribution of the scattered neutron is recorded using Legendre coefficients (the rejection method is a powerful technique for sampling the angle distribution that is described by a Legendre polynomial function); (4)the truncated exponential pdf, which is used to force the neutron to interact in a given sampled region (it is optional in the CEARCPG code); (5)the splitting method, which is used to increase the number of prompt gamma rays or gamma rays from the spontaneous fission neutron source; (6) a Monte Carlo generated detector response function, which is used to transform the incident gamma rays on the detector to a pulse-height spectrum. This latter technique is considered to be very

powerful and can save as much as 50% of the total tracking time for each gamma-ray history in the Monte Carlo simulation. The detector response functions are usually very accurate since the Monte Carlo simulation code with which the detector response functions are calculated contains very accurate semi-empirical models.

2.1 Gamma-ray sampling

There are two main sources of the gamma rays produced in PGNAA analysis. One is from the neutron inelastic scattering reaction and the other is from the neutron radiative capture reaction. The commonly used method for generating prompt gamma rays in Monte Carlo codes is to sample from a pre-calculated gamma-ray table - the MCNP code does this. This method can only be used to predict single spectra since there is no information on the coincidence relationship among the gamma rays. Fortunately, ENSDF data contains information that can be used to sample prompt gamma rays in coincidence. This includes the energy, relative intensity of each prompt gamma rays as well as structure information on each. Combining this with ENDF or JENDL data, one can determine the starting level of the scheme. For the neutron inelastic scattering reaction, the cross sections of each excited level are given by ENDF or JENDL library which are represented by MF=3, MT=51-90. Flag LR shows the de-excitation type of each excited level. If LR=0, it means the excited level decays by emitting a photon. Only the levels which decay by emitting photons are considered in order to increase computation performance. The weight of the neutron and the gamma ray that is produced is adjusted based on the excited level cross section. When the excited level is sampled, the nucleus structure scheme is employed to sample the prompt gamma rays. For the neutron radiative capture reaction, the energy of the highest excited level of the product nucleus is the Q value of reaction. Figure 1 shows the typical scheme of the ^{13}C nucleus produced by the ^{12}C (n,γ) reaction.

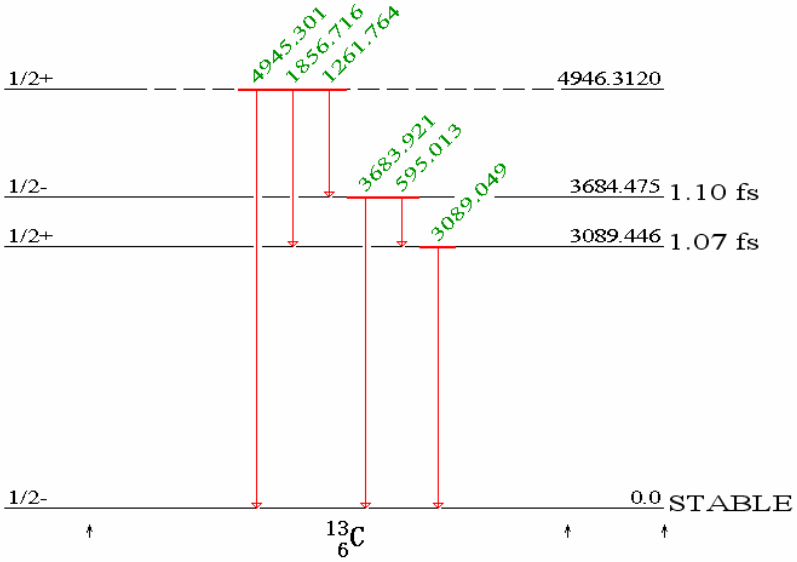

Figure 1. ^{13}C nucleus scheme of the ^{12}C (n,γ) reaction

For the ^{12}C(n, γ) ^{13}C reaction, the Q value is 4.946 MeV, equaling the energy of the highest excited level of the ^{13}C nucleus. Prompt gamma rays can be sampled from the highest level. Because the life time of the excited level is several femtoseconds, sequential gamma rays from these levels can be considered to be in coincidence. Sampling gamma rays generated by the

neutron inelastic scattering reactions more complex than sampling from the neutron radiative capture reaction. The ENDF data must be used to determine which level will be excited by the neutron inelastic scattering reaction.

3 CEARCPG CODE BENCHMARK

Several experiments have been done to benchmark the CEARCPG code.

3.1 Bulk coal sample

The geometry schematic of the bulk coal sample measurement is plotted in Figure 2. The geometry is plotted by using the Visual Editor of MCNP5. The prompt gamma rays are measured by a 5" X 5" NaI detector. The coal sample contains 17 elements. Since there is only one NaI detector, this experiment can be used to validate the capability of single spectra simulation of the CEARCPG code. Another simulation is also carried out by using MCNP5 for comparison. The measured spectra and the simulation results are plotted in Figure 3.

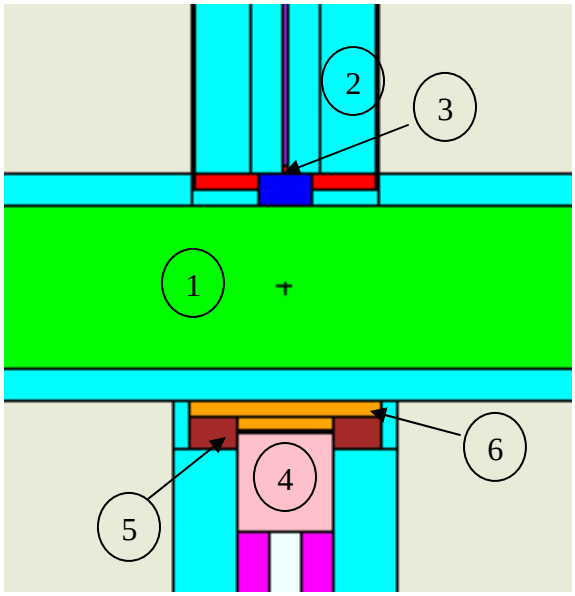

Figure 2. Schematic of ETI prototype, where (1) is coal sample, (2) is polyethylene, (3) is ^{252}Cf neutron source, (4) is NaI detector, (5) is lead shielding, and (6) is Lithium loaded polyethylene

The simulated spectra are normalized at the Nitrogen peak. Compared to the MCNP simulation results, the result of the CEARCPG code is closer to the experiment since the CEARCPG code also considers the fission gamma rays of ^{252}Cf neutron source, NaI detector activation gamma rays [13] and the gamma rays from the natural background. The code CURLLS [14] developed by CEAR is used to fit the coal sample. The fitting result is plotted in Figure 4. The Reduced Chi-Square value is 23; this is a statistic used to check the fitting goodness and is defined by Equation 1.

$$\chi^2 = \frac{1}{v}\sum_{n1}^{n2}\frac{e_i^2}{\sigma_i^2} \qquad (1)$$

Where σ_i is the standard deviation of each channel, v is the number of degrees of freedom, e_i is the error in channel i, n_1 and n_2 are the starting and ending channel numbers of fitting.

3.2 Pure sulfur sample

The geometry schematic for the pure sulfur sample measurement is shown in Figure 5. This experiment is designed to benchmark the capability of the coincidence spectrum simulation of the CEARCPG code. Two 6" X 6" NaI detectors were used to measure the single spectra and coincidence spectra of a pure sulfur bulk sample.

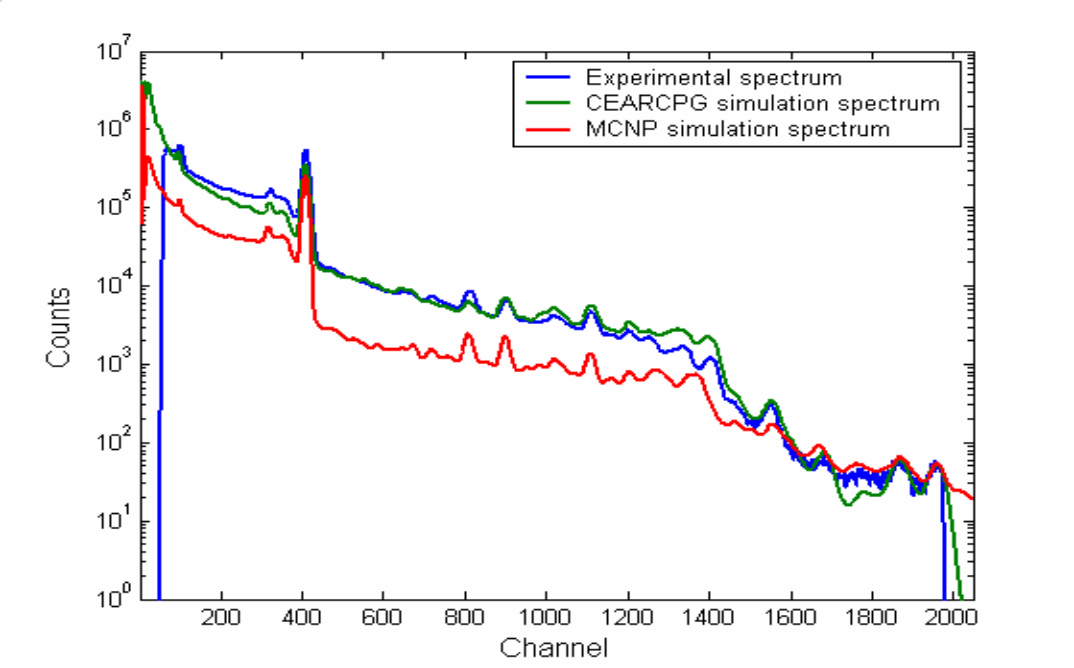

Figure 3. The measured spectrum and simulation spectra of coal sample

The measured spectrum and the fitting spectrum are plotted in Figure 6. The coincidence spectrum is recorded as a two-dimensional spectrum. One energy window is outlined that corresponds to the Q value of the ^{32}S (n, γ)^{33}S reaction [11], 8.64 MeV. The energy window is applied to both the experimental coincidence spectrum and the simulated spectrum. The blue line in Figure 6-3 is the spectrum obtained by projecting the experimental spectrum in the window to the X axis.

Compared to the single spectrum, the Q-value summing projection spectrum has much better peak resolution and signal-to–noise ratio. This is the biggest advantage of the coincidence technique over single energy measurement. Two pairs of peaks can be observed in the Q value diagonal summing spectrum, 0.84 and 7.80, 3.22 and 5.42 MeV, which are unique for every isotope. This feature shows that these "characteristic prompt gamma rays" can be used to identify the isotope and to determine their concentration. The green spectrum is the fitting result

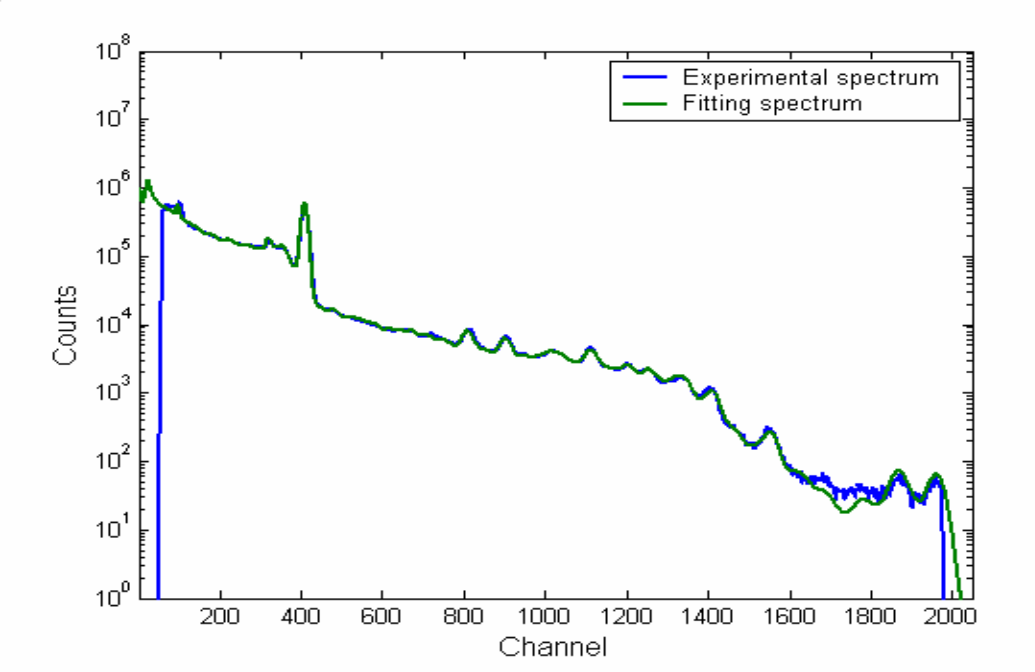

Figure 4. The fitting results of coal sample. The fitting result is obtained by using code CURLLS with the library spectra generated by the CEARCPG code.

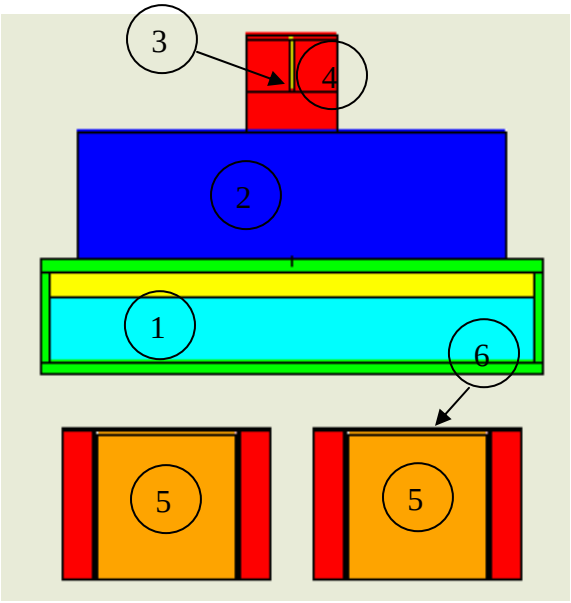

Figure 5. The scheme of ETI prototype, where (1) is Sulfur sample. (2) is wax, (3) is ^{252}Cf neutron source, (4) is lead shield, (5) is NaI detector, (6) is Lithium polyethylene

by using the CURLLS code with the simulation Q value diagonal summing spectrum. The fitting spectrum fits the experimental spectrum very well and gives a Reduced Chi-Square of only 2.1.

The difference is probably caused by chance coincidence in the experimental measurement, which cannot be predicted by the CEARCPG code at present.

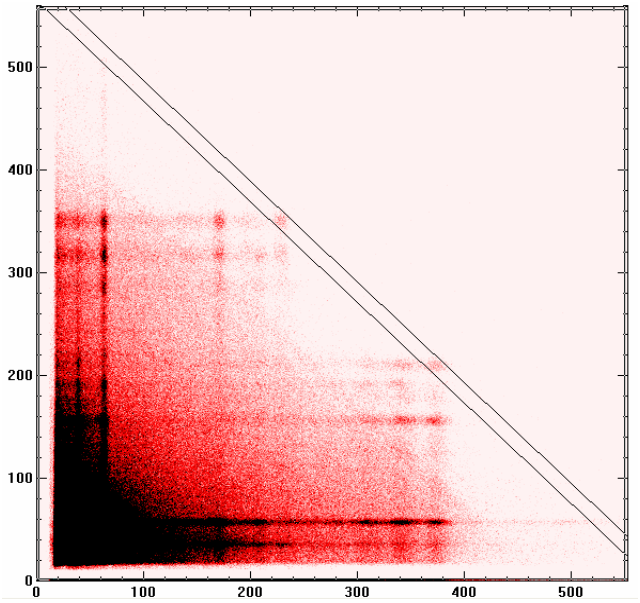

Figure 6-1

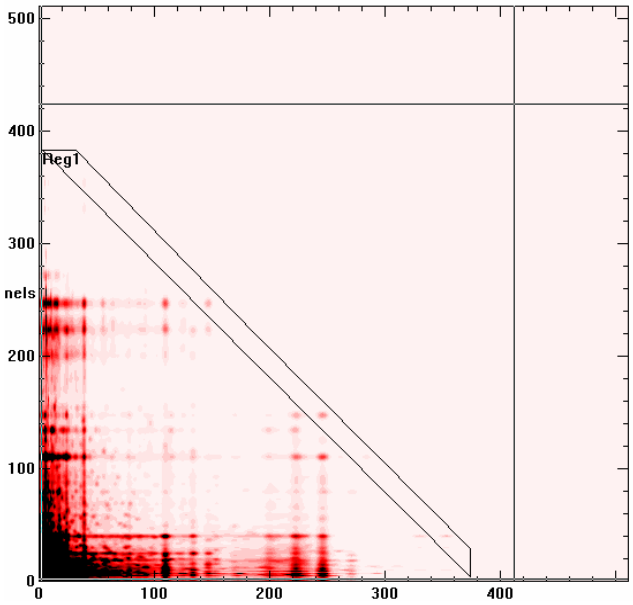

Figure 6-2

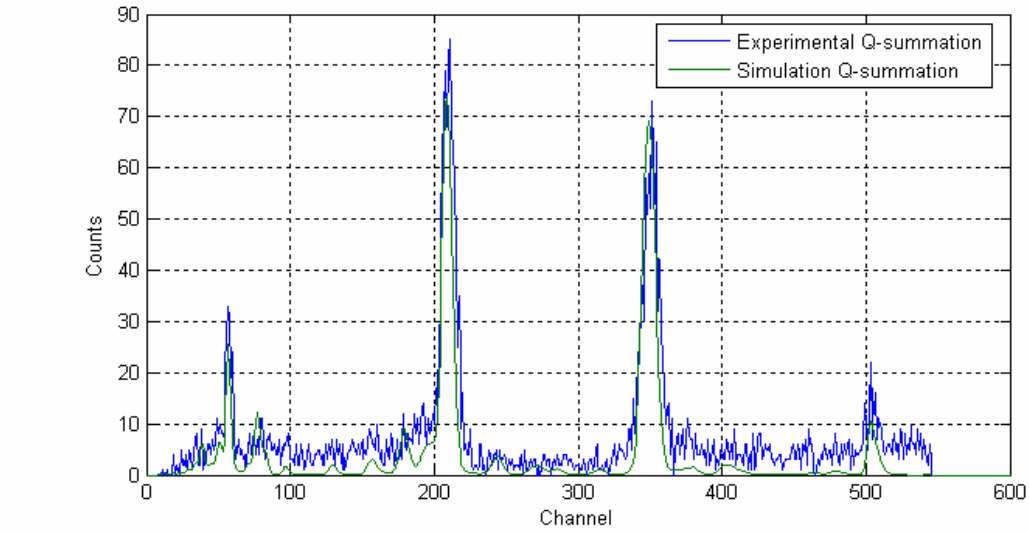

Figure 6-3

Figure 6. The two dimension coincidence spectra of a pure sulfur sample. Figure 6-1 is the experimental coincidence spectrum recorded as a two-dimensional spectrum ; Figure 6-2 is the simulated coincidence spectrum and Figure 6-3 shows the comparison between the experimental spectrum and the fitting spectrum for the Q-value diagonal summing spectrum

4 CONCLUSIONS

Feasibility studies by several researchers [8, 9,12] have been undertaken to prove that the coincidence technique can be applied to PGNAA analysis and it can improve its accuracy. In this

work, the CEARCPG code is used to predict the single and coincidence spectra of bulk coal and pure sulfur samples. The results show that the simulation results are accurate and the code will be useful for optimizing the coincidence PGNAA application. The primary disadvantage of coincidence PGNAA is its low intensity. It is one of the objects of the CEARCPG code to optimize the design of the coincidence PGNAA application in order to increase the coincidence counts. The geometry arrangement for the next simulation is shown in Figure 7. In this setup, one 6" X 6" NaI detector is surrounded by six 3" X 3" NaI detectors which serve as a "trigger". Theoretically, the large surface NaI detectors can increase the coincidence counts.

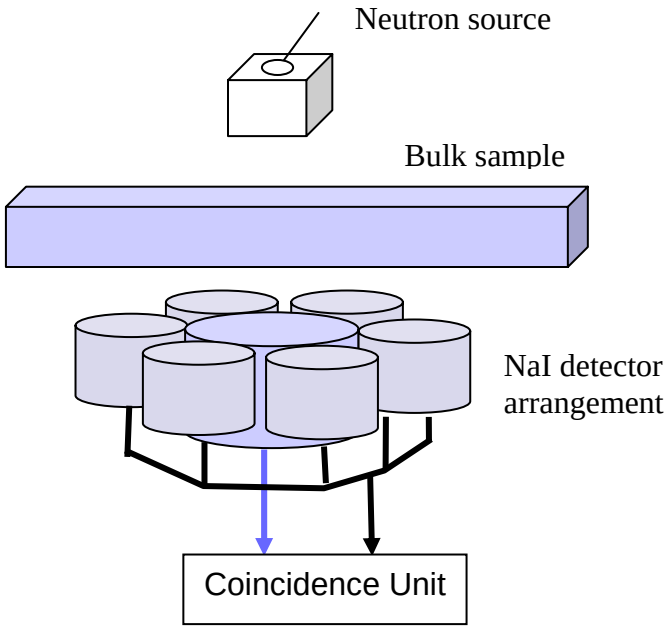

Figure 7. The geometry setup for coincidence PGNAA application optimization

5 ACKNOWLEDGMENTS

The authors gratefully acknowledge the Associates Program for Nuclear techniques in Oil Well Logging with present members Baker Atlas, ExxonMobil, Shell, and Computalog/Advantage Engineering for their support. We also acknowledge the U.S. Department of Energy for both an STTR grant with Sabia Inc. and a Nuclear Engineering Educational Research grant (Grant Nos. DE-FG03-02ER86154 and DE-FG07-02ID14330).

6 REFERENCES

[1]. T. Gozani, E. Elias, V. Orphan, J. Reed, and D. Shreve, "Prompt – Neutron Activation Analysis – Applications to Coal Analysis", *Trans. Am. Nucl*. Soc. **26**, pp.160, (1977)

[2]. G. Reynolds, H. Bozorgmanesh, E. Elias, T. Gozani, T.Maung, anf V. Orphan, "Nuclear Assay of Coal- Volume I, Coal Composition by Prompt Neutron Activation Analysis – Basic Experiment", *Report EPRI EP-989*, **Vol. 1** (1979)

[3]. C.M. Shyu, T. He, K. Verghese, and R.P. Gardner, "Monte Carlo-library least-squares principle for nuclear analysers". *Transactions of the American Nuclear Society,* Suppl. **3**, **56**, 4446. (1988.)

[4]. J.H. Marshall III and J.F. Zumberge, "on-line Measurements of Bulk Coal Using Prompt Gamma Neutron Activation Analysis", *nucl. Geophys.* **3(4)**, pp.445, (1989)

[5]. 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", *Nuclear Geophysics,* **7(2)**, pp.241-268 (1993)

[6]. Wenchao Zhang and Robin P. Gardner, "The analog linear interpolation approach for Monte Carlo simulation of PGNAA: The CEARPGA code", *Nuclear Instruments and Methods in Physics Research B,* **213**, pp.116-123 (2004)

[7]. R.P. Gardner, C.W. Mayo , E.S. El-Sayyed , W.A Metwally,, Y Zheng, M Poezart,. "A feasibility study of a coincidence counting approach for PGNAA applications". *Appl. Radiat. Isot*. **53**, 515–526. (2000)

[8]. P.P Ember, T. Belgya, G.L. Molnár, "Improvement of the capabilities of PGAA by coincidence techniques". *Appl. Radiat. Isot*. **56**, pp.535–541. (2002)

[9]. P.P. Ember, T. Belgya, J.L. Weil, G.L. Molnár, "Coincidence measurement setup for PGAA and nuclear structure studies". *Applied Radiation and Isotopes.* **57**, pp. 573-577, (2002)

[10]. A.H. Wapstra, "Alpha-, Beta- and Gamma-ray Spectroscopy", *5th Edition*, North-Holland Publishing Company, Amsterdam, New York, Oxford, **Vol.1** pp 539-555 (Chapter VIII/C). (1979)

[11]. W.A. Metwally, R.P. Gardner*, C.W. Mayo, "Two-dimensional diagonal summing of coincidence spectra for bulk PGNAA applications", *Nuclear Instruments & Methods in Physics research.* **A 525** pp.511-517 (2004)

[12]. Walid A. Metwally, Robin P. Gardner, Charles W. Mayo, "Elemental PGNAA analysis using gamma–gamma coincidence counting with the library least-squares approach", *Nuclear Instruments and Methods in Physics Research Section B.* **Volume 213**, pp. 394-399. January 2004,

[13]. Robin P. Gardner, El. Sayyed, Yuanshui Zheng, Stephanie Hayden, Charles W. Mayo, "NaI detector neutron activation spectra for PGNAA application", *Applied Radiation and Isotopes*. **53** pp. 483-497, (2000)

[14]. R.P. Gardner, A. Sood, Y. Y. Wang, L. Liu, P. Guo, and R.J. Cehrek, “Single Peak Versus Library Least-Squares Analysis Method for PGNAA Analysis of Vitrified Waste”. *Appl. Radiat. Isot.* **Vol. 48**, No. 10-12, pp. 1331-1335, (1997)