FINAL REPORT

For

PROMPT GAMMA-RAY NEUTRON ACTIVATION ANALYSIS
(PGNAA) FOR ELEMENTAL ANALYSIS

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ABSTRACT

This research project was to improve the prompt gamma-ray neutron activation analysis (PGNAA) measurement approach for bulk analysis, oil well logging, and small sample thermal neutron beam applications. This is accomplished by developing a Monte Carlo code called CEARCPG that describes both normal and coincidence PGNAA counting. The latter has the potential to greatly reduce the background of these devices and, therefore, increase the signal-to-noise ratio (S/N) of this measurement approach. This code is the first that is capable of describing coincidence PGNAA counting. The code is benchmarked with experiments that are similar to the applications of interest and is then used to simulate and optimize the applications of interest. Since all elemental coincidence schemes are not known, one of the tasks is to determine the schemes of elements of importance. Another task is to improve the Monte Carlo – Library Least-Squares (MCLLS) approach, which was invented and developed at CEAR. Finally, the MCLLS approach and other inverse analysis approaches are to be demonstrated for coincidence PGNAA.

All these tasks have been accomplished and it appears that the coincidence PGNAA approach will be very beneficial in the three applications investigated of bulk analysis, oil well logging, and small sample thermal neutron beam analyses.

INTRODUCTION

A previous NEER grant (Award Number DE-FG07-99ID13775) entitled “Coincidence Prompt Gamma-Ray Neutron Activation Analysis” led to the present grant. The present grant builds on the previous one which established feasibility of the coincidence counting approach.

A large number of technical papers (37) and four PhD theses have been published or accepted for publication that were funded by these two grants. They are listed in the References Section and give more detail than the present report. These two grants have supported work on a wide range of topics that are pertinent to normal and coincidence PGNAA. This includes: (1) work on pulse pile-up that allows one to correct for the spectral distortion caused by high counting rates, (2) the development and benchmarking of Monte Carlo models capable of describing both normal and coincidence PGNAA, (3) the development and testing of inverse analysis methods for PGNAA and CPGNAA including the Monte Carlo – Library Least-Squares (MCLLS) approach, (4) the investigation of prompt gamma-ray elemental coincidence decay schemes, and (5) practical application to bulk analysis, oil well logging, and small sample thermal neutron beam analysis.

OBJECTIVE 1 RESULTS

Objective 1 (25% of Total Effort) was to continue to modify our existing CEARPGA Monte Carlo code to include coincidence PGNAA (CPGNAA). The modified code would be named CEARCPG.

This objective has been completely accomplished. The PhD thesis of Xiaogang Han (Han, 2005) gives the details of the CEARCPG code. A paper has been prepared and submitted (Han, Gardner, and Metwally, 2006) that has been accepted for publication in Nuclear Science and Engineering.

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 coincidence spectra for prompt gamma-ray neutron activation analysis (PGNAA). The user needs to create the input file that will be read by CEARCPG. The information in the input file contains: (1) the geometry specification; (2) the description of materials; (3) neutron source information; and (4) 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 Monte Carlo - Library Least-Squares (MCLLS) fitting.

The main features of the CEARCPG code are:

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

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

- (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, and
- (c) The neutron inelastic scattering libraries. This library is used to sample the gamma rays that are produced through the neutron inelastic scattering reaction.

2. The library for gamma rays include the elements Z=1 to 100, which comes from the EPDL97 library. The gamma-ray energy range is from 0.01 to 20 MeV.

3. Most of the CEARCPG code input cards take the same form as those used in MCNP. 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 nuclides structure data are the same as the ENDF and ENSDF nuclear data format. In MNCP, the nuclear data must be processed into the desired format by using the NJOY code. It is easy for users to update. For CEARCPG code usage, any new nuclear data is copied directly without having to conform to a specific data format.

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

1. The stratified sampling technique. It 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. It 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. The truncated 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. By using this technique, 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.

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

In prompt gamma-ray neutron activation analysis, photons are generated from neutron inelastic or neutron capture reactions. For most (if not all) of the existing Monte Carlo codes, such as MCNP, photons are sampled from pre-calculated 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 is 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 which 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 which is produced from the $^{12}\text{C}(n,\gamma)$ reaction. From that scheme, the following information is supplied for prompt gamma-ray sampling. This is: (1) the energy and intensity of each prompt gamma ray; (2) the energy of each excited level; (3) the life time of each excited level and; (4) 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 de-excite 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 only considers the case of photon emission. If there is some internal conversion from a given level, the photon weight needs to be adjusted. Since the life time 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 is used first to determine which level will be sampled. This cross section data is from the ENSDF nuclear data library. When the excited level is determined, the nuclei structure data for the neutron inelastic scattering reaction is used to sample the appropriate gamma rays.

Compared to the algorithm used in the Analog Linear Interpolation 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.

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 is now more complete.

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 not only emits fission neutrons, but also emits 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 are from the experimental work by Verbinski. Both neutrons and photons are emitted isotropically. Since there is more than one fission gamma ray emitted per spontaneous fission, the multiplicity of fission gamma rays are also taken into account. These fission gamma rays might contribute to the coincidence spectrum.

The CEARCPG code only considers neutron elastic scattering, neutron inelastic scattering, and the neutron capture reaction, which are the major reactions for non-fissionable 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.

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 spectra 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.

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 only takes into account photoelectric effect, Compton scattering, and pair production interactions. There is no X-ray and 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 reaction,
3. the neutron source fission reaction (a background source), and
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 will be convoluted with the detector response function to convert the incident photon flux tally into a pulse-height spectrum tally.

OBJECTIVE 2 RESULTS

Objective 2 (10% of Total Effort) was to benchmark the CEARCPG code with selected experiments that are representative of the bulk analysis and oil well logging (C/O ratio) CPGNAA applications and neutron beam analysis for small samples.

This objective has been completed entirely. Benchmark results for several coals have been made for the bulk analysis application, benchmark results have been made for two new prototype C/O logging tools, and thermal neutron beam benchmarks have been made for Hg and Gd samples. The prototype PGNAA analyzer is shown in Figure 2. The experimental and CEARCPG simulated coal spectra are shown in Figure 3 and illustrate the accuracy of the code. Six coals of known sulfur composition were used and the code simulated values matched very well.

Prototype C/O device results have also been obtained as have small sample results in the thermal neutron beam. These results are also quite accurate and will be shown later as part of Objective 5.

Figure 2. The geometry configuration of the ETI prototype, where 1 is coal sample, 2 the polyethylene, 3 bismuth with the neutron source, 4 air, 5 aluminum, 6 Lithium polyethylene, 7 lead, 8 NaI detector, 9 SiO₂ (PMT) and 10 is paraffin

Figure 3. Experimental and CEARCPG spectra for a coal sample.

Figure 4. Experimental coincidence arrangement for Sulfur sample.

Figure 5. The measured single and total coincidence spectra of a pure sulfur sample.

OBJECTIVE 3 RESULTS

Objective 3 (30% of Total Effort) was to continue to obtain coincidence decay schemes using 3D spectroscopy of the elements of primary interest in bulk analysis (primarily coal and cement) and oil well logging (limestone and sandstone formations).

This objective has been accomplished to the extent that sulfur, mercury, and gadolinium have been studied. The results for sulfur are shown in Figures 6 and 7. Figure 6 shows the 2D spectrum obtained for a pure sulfur sample with the Q-value diagonal window superimposed on it. Figure 7 shows the counts in the diagonal window that are from one of the detectors for both the experimental and CEARCPG predicted cases. It is obvious that the results are quite good.

Figures 8 and 9 show the same results for a mercury sample.

Figure 6. The Q-value diagonal summation window for pure Sulfur sample.

Figure 7. The experimental and CEARCPG predicted projection spectra of the Q-value diagonal summation for a pure Sulfur sample.

Figure 8. Schematic diagram of the mercury experiment.

Figure 9. The experimental Q-value projection spectrum of a pure mercury sample.

Figure 10. Gadolinium singles and coincidence spectra.

Table 1. Coincidence Pairs Of Gd-157.

Coincidence Pair	Coincidence Intensity(per 10000 captures)
6750.0 kev ----1187.1 kev	126
6750.0 kev ----1107.61 kev	148
5903.0 kev ---- 1010.0 kev ---- 944.2 kev	58
5903.0 kev-----875.0 kev ----- 895.5 kev----181.9 kev	46
5903.0 kev-----768.4 kev ----- 1186.0kev	33
181.9 kev ---- 79.5 kev	436
897.6 kev ---- 79.5 kev	70
944.2 kev ---- 79.5 kev	204
962.1 kev ----- 79.5 kev	130
1107.7 kev -----79.5kev	107
1186.0 kev ----79.5 kev	102
277.6 kev ---- 181.9 kev	113
780.2 kev ----- 181.9 kev	250
897.5 kev ----- 181.9 kev	327
915.0 kev ----- 181.9 kev	107
1097.0kev ---- 181.9 kev	180
1119.0 kev ----- 181.9 kev	312
1141.5kev -----181.9kev	170

The Q-value for Gd-157 is 7.937 Mev. From the above table, it is seen that there are a lot of coincidence pairs for 79.5kev and 181.9kev gamma rays. The 79.5kev one is too low for a scintillation detector to detect. Therefore, the coincidence with 181.9kev is our main concern. There is no one specific high energy gamma ray (above 3Mev) which is in coincidence with 181.9kev and is much more intense than any other high energy gamma ray. The fact is that neutron captured gamma rays of Gd are very complicated. There are a lot of high energy gamma rays which are in coincidence with the 181.9kev gamma ray to make the sum equal the Q-value, but no one of them is more intense than any other.

OBJECTIVE 4 RESULTS

Objective 4 (10% of Total Effort) was to continue to develop the Monte Carlo – Library Least-Squares (MCLLS) approach to the coincidence nonlinear inverse analysis problem for PGNAAs bulk analysis and oil well logging.

This work has been carried out as an extension of the use and development of normal PGNAAs for both of these applications. The normal MCLLS is discussed in the section on Objective 2 Results. Figure 3 illustrates how well this approach can be used for the bulk analysis (coal) application. Two recent papers have been prepared on this topic for these two applications (Han, Gardner, Metwally, and Guo, 2006 and Gardner, Han, and Guo, 2006). These papers discuss possible variations in the MCLLS approach when two-dimensional coincidence spectra are accumulated. The proposed optimum approach for the coincidence counting method is discussed in the recent paper by Metwally, Mayo, Han, and Gardner (2005).

OBJECTIVE 5 RESULTS

Objective 5 (25% of Total Effort) was to investigate various practical methods for applying CPGNAAs to (a) bulk analysis, (b) oil well logging, and (c) small sample neutron beam analysis. Methods so far identified include: (1) use of multiple and/or large low resolution detectors as the *trigger* detectors for a and c, (2) use of ^{241}Am -Be neutron sources (with their very long half-life) for a and possibly b, and (3) use on angular correlation specifics for a, b, and c.

Some work has been done in this area, but much more needs to be done. This is the kind of objective that will require many years to complete. The most recent work on this objective is given in the two papers by Han, Gardner, Metwally, and Guo (2006) and Gardner, Han, and Guo (2006).

DISCUSSION, CONCLUSIONS, AND FUTURE WORK

The grant reported on here has supported or partially supported four PhD students (W. Zhang, W.A. Metwally, W. Guo, and X. Han). The last three of these were also supported as Post-Doctoral students after graduation. This provided needed continuity in this research.

The original idea was to investigate coincidence counting to minimize the large amount of background in the normal prompt gamma-ray neutron activation analysis (PGNAA) spectral response. After some initial experiments with ordinary coincidence counting equipment it occurred to us that it would be nice to have a coincidence system that is capable of obtaining a more complete set of data that would include the complete two-dimensional coincidence spectra in each of the two detectors that are used. With that in mind we purchased a Sparrow system for that purpose. Obtaining data with this system enabled us to have the greatest amount of flexibility in treating the data. This type of system is routinely used by nuclear physicists for determining coincidence decay schemes, but to our knowledge has not been used for engineering measurements like bulk analysis or oil well logging with PGNAA devices.

We found that we could essentially eliminate the background from PGNAA devices for the bulk analysis problem. Typically the background in these devices represents 70 to 85 % of the total spectral response. The signal from hydrogen is also essentially eliminated, which turns out to be good in the sense that it gives a signal about two orders of magnitude higher than all other elements present in coal. Eliminating the hydrogen response (2.223 MeV prompt gamma ray) allows low energies to be used – such as the most prominent sulfur prompt gamma-ray energy of 0.841 MeV. Normally commercial suppliers of PGNAA devices for bulk analysis of coal suggest not using prompt gamma rays below about 2.3 MeV.

Unfortunately, when the background is reduced the signal is also reduced when two normal sized detectors are used. So the accuracy is not necessarily increased when two normal sized detectors are used for coincidence counting. To combat this problem we suggest that one very large detector (which can have much poorer resolution than normal) be used as a trigger detector for the coincidence counting. This results in the signal not being reduced as much and gives better accuracy. This is treated mathematically in detail in the paper by Gardner, Metwally, Zhang, Han, and Mayo (2003).

Another practical implication of these results is that typical commercial PGNAA devices use the neutron source ^{252}Cf which has a half life of only 2.55 years. This source is used rather than an $^{241}\text{Am-Be}$ source because the latter has considerably more background gamma rays. With the coincidence approach

one can use the ^{241}Am -Be source with its half-life of 458 years and avoid having to replace the neutron source every five years or so.

A number of other improvements have been made during the course of this research including: (1) pulse pile-up treatments which allow the elimination of the distortion due to pulse pile up at high counting rates and (2) improvements in the Monte Carlo model used to generate elemental libraries for the MCLLS inverse analysis approach for both normal and coincidence PGNAAs. These improvements are described in detail in the references.

The future work anticipated includes the investigation of alternate inverse algorithms for the coincidence PGNAAs analysis such as the Q-value summing or similar techniques and the optimization of coincidence devices for various applications.

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