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The Monte Carlo code CEARCPG for coincidence prompt gamma-ray neutron activation analysis

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Abstract

Prompt gamma-ray neutron activation analysis (PGNAA) is widely used to determine the elemental composition of bulk samples. The detection sensitivities of PGNAA are often restricted by the inherent poor signal-to-noise ratio (SNR). There are many sources of noise (background) including the natural background, neutron activation of the detector, gamma-rays associated with the neutron source and prompt gamma-rays from the structural materials of the analyzer. Results of the prompt gamma-ray coincidence technique show that it could greatly improve the SNR by removing almost all of the background interferences. The first specific Monte Carlo code (CEARCPG) for coincidence PGNAA has been developed at the Center for Engineering Application of Radioisotopes (CEAR) to explore the capabilities of this technique. Benchmark bulk sample experiments have been performed with coal, sulfur, and mercury samples and indicate that the code is accurate and will be very useful in the design of coincidence PGNAA devices.

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Keywords: Coincidence; PGNAA; Monte Carlo; CEARCPG

1. Introduction

Prompt gamma-ray neutron activation analysis (PGNAA) is a nondestructive, rapid analytical technology and it can be used on bulk material streams. It uses the gamma-rays emitted during neutron irradiation. However, the prompt gamma-ray spectrum has a low-signal-to-noise (S/N) ratio. Primarily the background interferences are from the neutron excitation source, prompt gamma-rays from the structural materials of the PGNAA device and activation of the NaI detector [1], which is widely used in the PGNAA application. A possible improvement is to change the traditional PGNAA detection technique and introduce the γ–γ coincidence technique [2,3] since all the prompt gamma-rays of all radioisotopes are emitted in coincidence except hydrogen. Research on the coincidence measurement started at the Center for Engineering Application of Radioisotopes (CEAR) several years ago. Several feasibility studies have already been done and show that this approach can effectively increase the S/N ratio and significantly depress the interferences. In this work, the coincidence PGNAA approach is carefully studied by using Monte Carlo method and all the simulated results are checked with experiments.

2. Monte Carlo simulation

Before this there was no existing Monte Carlo code that could be used to simulate prompt gamma-ray coincidence spectra. The commonly used method to generate prompt gamma-rays in Monte Carlo codes, such as MCNP, was to sample the prompt gamma-rays from the pre-calculated gamma-ray table for every photon-producing neutron interaction. The disadvantage of this method is that the relationship information is lost even when we know the energy and intensity of each prompt gamma-ray. The algorithm limits the existing Monte Carlo code to be used for

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coincidence simulation. A totally new algorithm is developed in the CEARCPG code. The basic idea of sampling the neutron-produced prompt gamma-rays is to sample the prompt gamma-rays from the excited scheme of the radioisotopes. Fig. 1 shows the typical scheme of the radioisotope ^{13}C which is produced through the $^{12}C(n,\gamma)^{13}C$ reaction. For neutron capture reactions, the energy of the highest level is the Q-value of the reaction. For this reaction, the Q-value is 4.946 MeV. The primary nuclear structure data is from the ENSDF nuclear data library [4] and the IEAE PGAA nuclear data library [5]. The typical ENSDF nuclear data have the information for each individual prompt gamma-ray, such as the energy, the relative intensity and the structure information. The structure

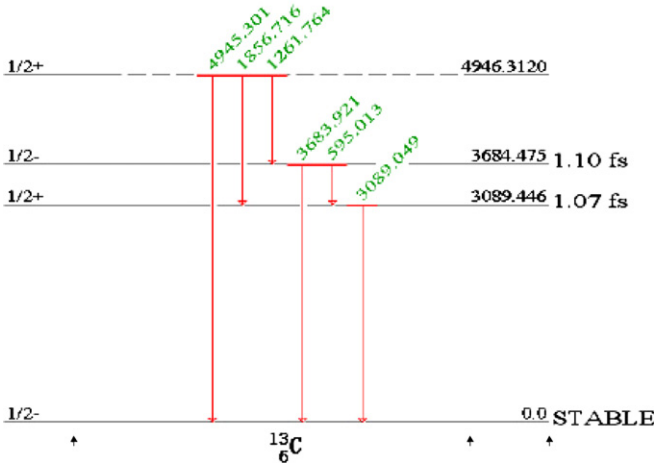

Fig. 1. The excitation scheme of ^{13}C produced by the $^{12}C(n,\gamma)^{13}C$ reaction.

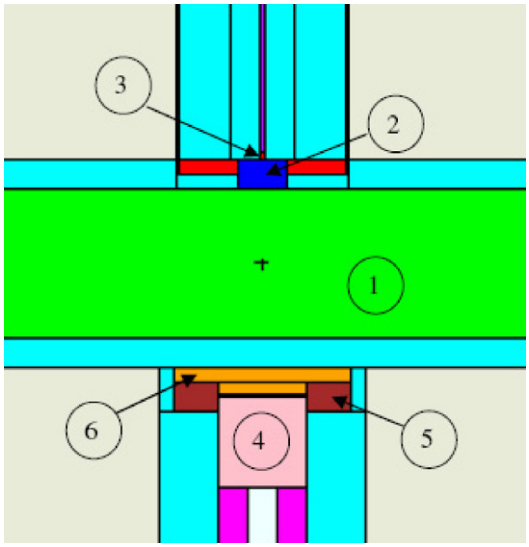

Fig. 2. Schematic diagram of the ETI prototype, where (1) is coal sample, (2) is polyethylene, (3) is the ^{252}Cf neutron source, (4) is the NaI detector, (5) is lead shielding and (6) is lithium loaded polyethylene.

information is important since it indicates at which level the prompt gamma-rays are generated from and which level it ends. Based on the scheme, the relationship among the prompt gamma-rays is completely known. This algorithm can be extended to sample the gamma-rays produced from neutron inelastic scattering reactions.

Code CEARCPG is a specific Monte Carlo code that treats an arbitrary three-dimensional configuration of user-defined materials bounded by first and second order surfaces. It can be used for neutron, photon, or neutron/photon transport. Both single and coincidence spectra are tallied at the end of the simulation. The generated

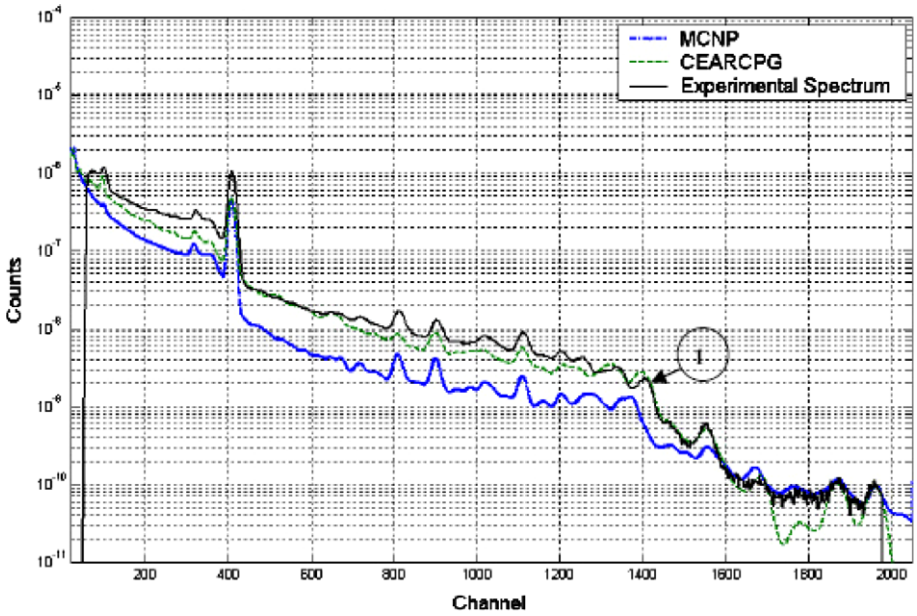

Fig. 3. The experimental and simulated spectra of a coal sample. Label 1 is the peak from the activation of the NaI detector.

elemental library spectra, both single and coincidence, are used by the Monte Carlo library least-square (MCLLS) approach [6] developed at CEAR. The main parts of code CEARCPG are listed as follows: (1) an initialization part which is used to define the geometry, required neutron cross sections, photon cross sections, angular distributions of scattered neutrons and the 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 with default neutron source ^{252}Cf and user-defined neutron source capabilities; (3) a neutron tracking part which is used to track the flight of neutrons and sample collision positions as well as neutron interaction types; (4) a prompt gamma-ray sampling part which is used to sample the prompt gamma-rays generated by neutron 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 singles/coincidence spectra of each element of interest.

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.
   (i) The neutron cross-section library, which is extracted from the ENDF/B-VI. 8300 K and the JENDL-3. 3300 K libraries.
   (ii) The (n,γ) scheme library, which is used to sample prompt gamma-rays caused by the (n,γ) reaction.
   (iii) The neutron inelastic scattering library. 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 $Z = 1$–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 takes 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.

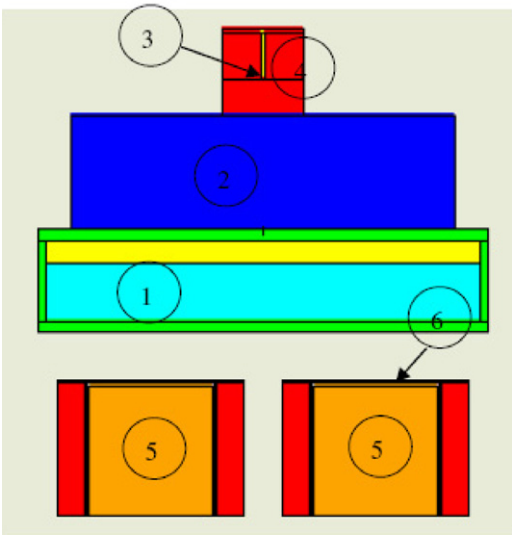

Fig. 4. The schematic diagram of the sulfur sample arrangement, where (1) is sulfur sample. (2) is wax, (3) is ^{252}Cf neutron source, (4) is lead shield, (5) is NaI detector and (6) is lithium polyethylene.

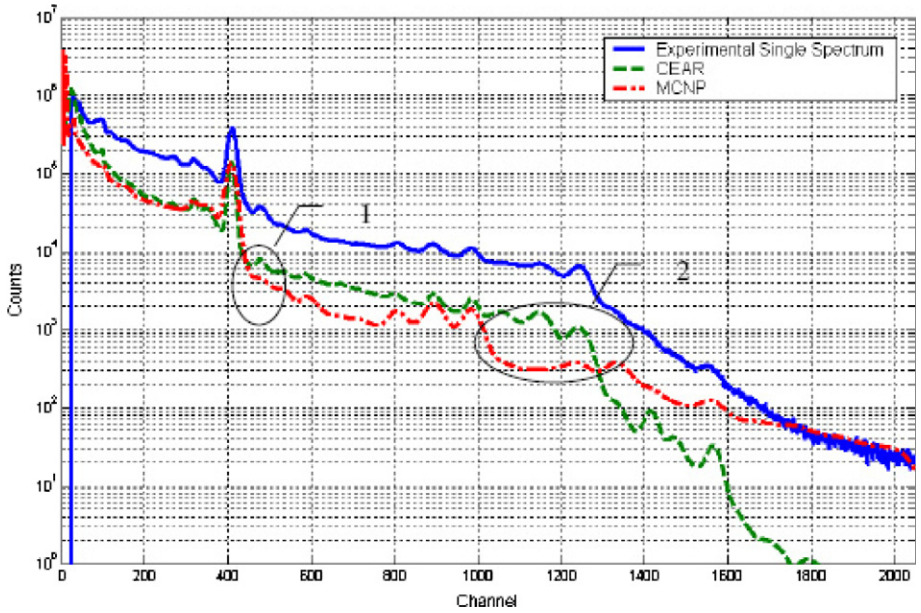

Fig. 5. The single spectra of a pure sulfur sample. Where region 1 is the gamma-rays from the natural background and region 2 is the gamma-rays from the NaI detector activation.

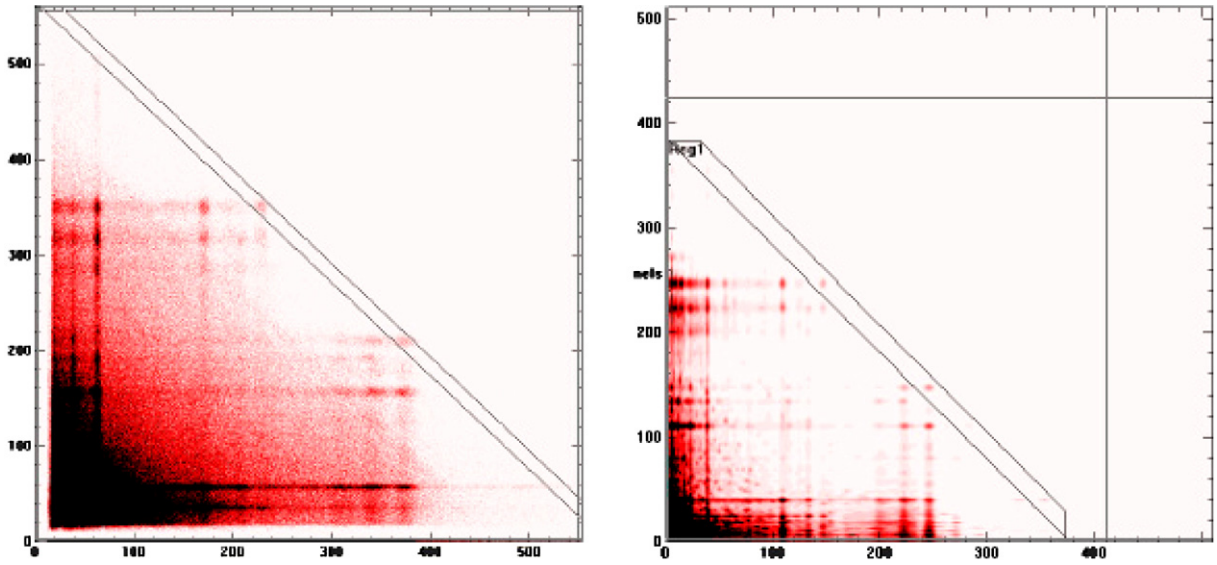

Fig. 6. The coincidence spectra of a pure sulfur sample. (a) the experimental coincidence spectrum; (b) the calculated coincidence spectrum.

The treatments of neutron transport are the same as those used in MCNP5. Only neutron capture reactions and neutron elastic/inelastic scattering reactions are taken into account. The (n,γ) reaction will be added in the future since it is important to oxygen for neutron inelastic scattering reaction. Angular distribution is considered for scattered neutrons based on ENDF nuclear data. The free gas model is applied for thermal neutron scattering. The treatments of photon transport are the same as the simple physics used in MCNP5. Only incoherent scattering, pair production and photo-electric reactions are considered in code CEARCPG.

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) 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 by 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; and (6) a Monte Carlo generated detector response function [7], 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

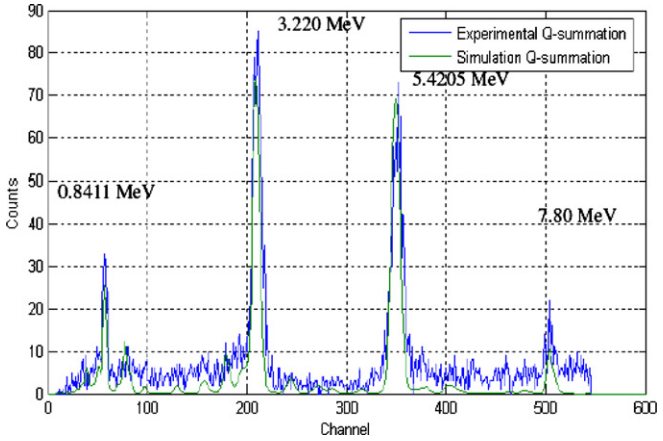

Fig. 7. The comparison between the experimental Q-value projection spectrum and the calculated spectrum.

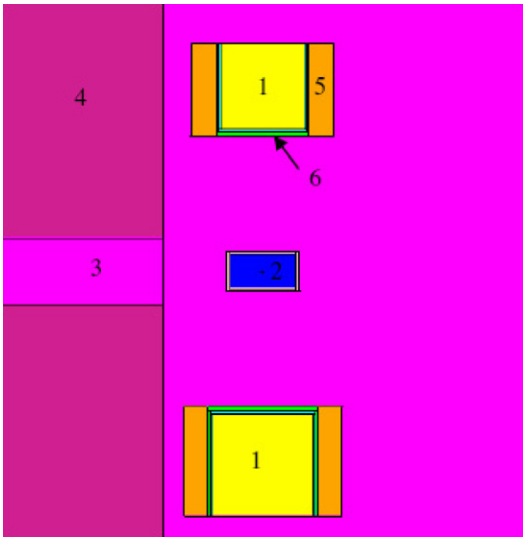

Fig. 8. The schematic diagram of the mercury sample arrangement, where (1) is NaI detector; (2) is mercury sample; (3) is the neutron beam; (4) is concrete, (5) is lead and (6) is lithium polyethylene.

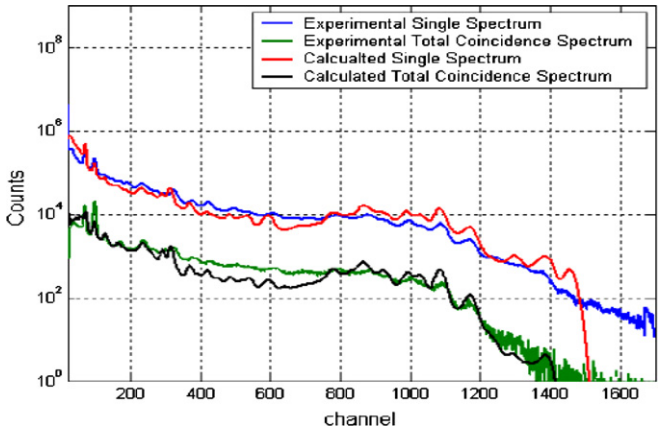

Fig. 9. The experimental spectrum versus the calculated spectrum of the mercury sample.

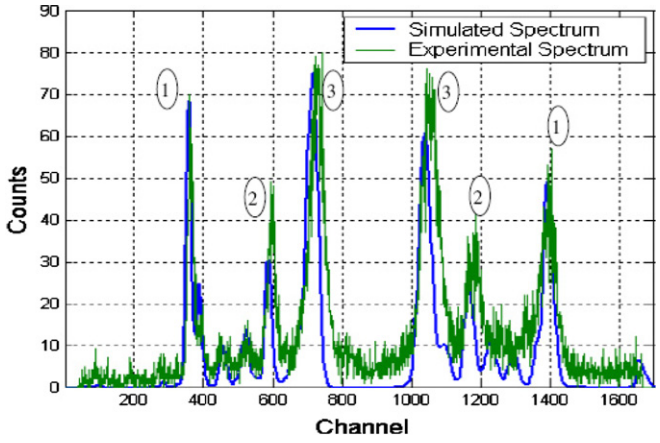

Fig. 11. The Q-value projection spectrum of the mercury sample.

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.

3. Benchmark experiments

3.1. Bulk coal sample

The simulation schematic is plotted in Fig. 2 by using the VisEd of MCNP5. One 5 in. × 5 in. NaI detector is used to measure the neutron-produced prompt gamma-rays. A simulation was also carried out with MCNP5 for comparison purposes. Since the ^{252}Cf spontaneous fission source not only emits fission neutrons, but also the fission gamma-rays, two separate simulations are done using MNCP5. The results are plotted in Fig. 3. All the spectra are normalized at the nitrogen peak. Compared to the spectrum of MCNP5, the spectrum of the CEARCPG code can correctly predict the spectrum produced from the activation of the NaI detector and is closer to the experimental spectrum. The computer time is 15.7 h on a Pentium IV 1.6 GHZ PC. The stratified sampling variance reduction technique guarantees that every elemental library spectrum has the same statistics.

3.2. Pure sulfur sample

Coincidence spectra are measured in this experiment by using two 6 in. × 6 in. NaI detectors. The schematic of the simulation is plotted in Fig. 4. The measured single spectrum and the calculated spectrum are plotted in Fig. 5. The two missed parts of the MCNP results are labeled.

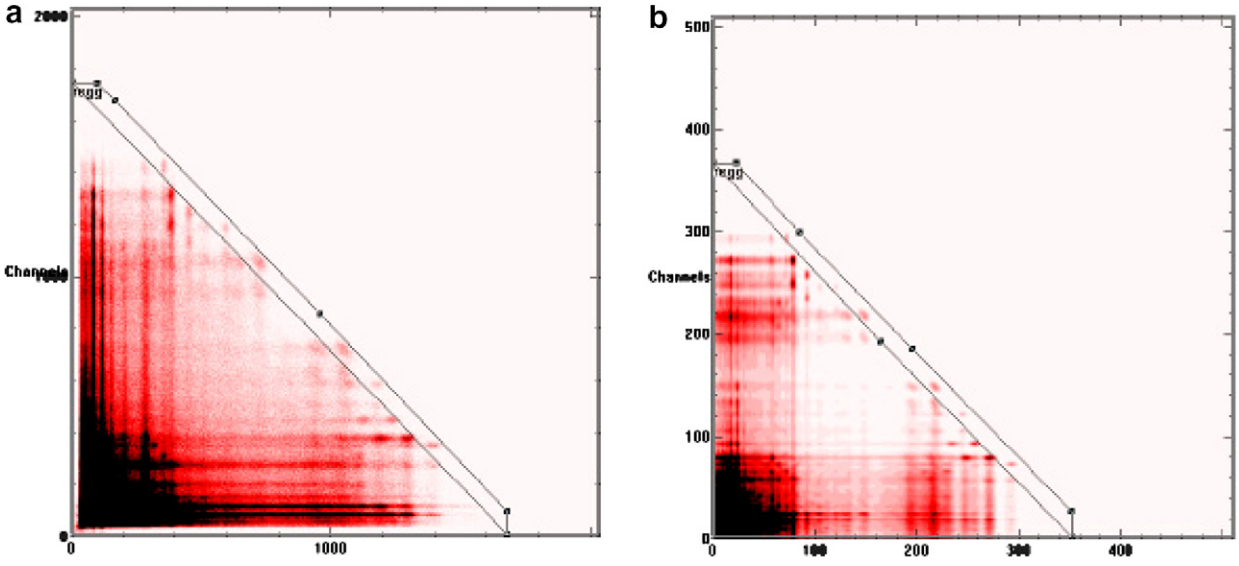

Fig. 10. The experimental two-dimensional spectrum versus the calculated two-dimensional spectrum of the mercury sample.

Table 1
Energies adding up to 8.028 MeV in the ^{200}Hg excitation scheme

Pair #	Energy in coincidence (MeV)
1	1.571 and 6.457
2	2.639 and 5.387
3	3.288 and 4.739
	3.185 and 4.841

Compared to the spectrum of CEARCPG, the spectrum of MCNP5 is better at the high-energy region. This is probably due to better statistics. The neutron history is 1E8 for the MCNP simulation and only 1E6 for the CEARCPG code. The computer time is 28 h on a Pentium IV 1.6 GHZ PC. Because the coincidence spectrum is recorded, a larger neutron history is needed than for the previous case. The coincidence spectrum is recorded as a two-dimensional spectrum. One energy window is outlined that corresponds to the Q-value of the $^{32}S(n,\gamma)^{33}S$ reaction, 8.641 MeV [8]. Both the experimental coincidence spectrum and the simulated spectrum are plotted in Fig. 6 and the Q-value projection is applied to both spectra. The projection spectra are plotted in Fig. 7. The results show that the calculated spectrum fits the experimental spectrum very well.

3.3. Pure mercury sample

This experiment is carried out in the thermal neutron beam of the NCSU PULSTAR Research Reactor. Two NaI detectors (5 in. × 5 in. and 6 in. × 6 in.) are arranged to face each other which are shown in Fig. 8. This case is simulated by using MCNP5 too. Unfortunately, the output spectrum of MCNP is not correct. The problem may come from the mercury data used in MCNP. The nuclear data of mercury used in MCNP5 is from ENDL92. The computer time for the CEARCPG code is 23 h. Only simulated spectra of code CEARCPG and experimental spectra are plotted in Fig. 9. Fig. 10 shows the experimental coincidence spectrum and the calculated coincidence spectrum. The Q-value projection is applied to the experimental coincidence spectrum and the calculated coincidence spectrum as well. The projection spectra are plotted in Fig. 11. The dominant coincidence pairs are labeled in the figure and their energies are listed in Table 1.

4. Conclusion

The CEARCPG code makes it possible to simulate the coincidence measurement of PGNAA. The results show that the CEARCPG code can be used to predict both the singles and coincidence spectra of coincidence PGNAA. The algorithm to generate the prompt gamma-rays used in CEARCPG could be implemented into MCNP and extended to simulate the decay gamma-rays of radioisotopes that are produced. In order to simulate the decay gamma-rays of radioisotopes, their decay schemes are needed. The CEARCPG code is useful for optimizing the coincidence application. Since the biggest disadvantage of the coincidence measurement is its low-counting rate, optimization is one of objectives of the CEARCPG code. Some optimization studies have already been done by using the CEARCPG code.

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