Nuclear Instruments and Methods in Physics Research A 652 (2011) 572–577

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Monte Carlo investigation and optimization of coincidence prompt gamma-ray neutron activation analysis

Jiaxin Wang*, Adan Calderon, Cody R. Peeples, Xianyun Ai, Robin P. Gardner

Center for Engineering Applications of Radioisotopes (CEAR), North Carolina State University, Raleigh, NC 27695-7909, USA

ARTICLE INFO

Available online 26 August 2010

Keywords:
Coincidence
PGNAA
Monte Carlo
CEARCPG

ABSTRACT

Normal Prompt Gamma-Ray Neutron Activation Analysis (PGNAA) suffers from a large inherent noise or background. The coincidence PGNAA approach is being investigated for eliminating almost all of the interfering backgrounds and thereby significantly improving the signal-to-noise ratio (SNR). This can be done since almost all of the prompt gamma rays from elements of interest are emitted in coincidence except hydrogen. However, it has been found previously that while the use of two normal NaI detectors greatly reduces the background, the signal is also greatly reduced so that very little improvement in standard deviation is obtained. With the help of MCNP5, the general-purpose Monte Carlo *N*-Particle code, and CEARCPG, the specific purpose Monte Carlo code for Coincidence PGNAA, further optimization of the proposed coincidence system is being accomplished. The idea pursued here is the use of a large area plastic scintillation detector as the trigger for coincidence events together with a normal large NaI detector. In this approach the detection solid angle is increased greatly, which directly increases the probability of coincidence detection. The 2D-coincidence spectrum obtained can then be projected to the axis representing the NaI detector to overcome the drawback of low energy resolution and photopeak intensity of the plastic scintillation detector and utilize the overall higher coincidence counting rate. To reach the best coincidence detection, the placement of detectors, sample, and the moderator of the neutron source have been optimized through Monte Carlo simulation.

1. Introduction

Prompt Gamma-Ray Neutron Activation Analysis (PGNAA) offers an on-stream, non-destructive, relatively rapid method for determination of elemental composition of bulk samples on conveyor belts. However, about 70–80% of the total PGNAA response is typically noise that comes from natural background (^{40}K, U, and Th), neutron excitation of detector prompt gamma rays, and gamma rays from the radioisotopes ^{24}Na and ^{128}I that are produced, gamma rays associated with the neutron source, and prompt gamma rays from the structural materials of the PGNAA devices. These sources of background highly limit the sensitivity and accuracy of PGNAA [1]. Also, hydrogen, both in the sample or non-sample region, is a common element that can increase the dead time and reduce the sensitivity of a PGNAA analyzer, due to the much higher magnitude of the hydrogen peak than the peaks of other elements of interest.

Thus, the coincidence PGNAA approach has been investigated to eliminate most of the interfering backgrounds and significantly improve the signal-to-noise ratio (SNR) since almost all of the prompt gamma rays from elements of interest are emitted in coincidence except for hydrogen. This preliminary approach has been validated by our research group in both experiments [2,3] and Monte Carlo simulation through a specific purpose code CEARCPG [4]. Unfortunately, in the initial arrangements studied the magnitude of the total coincidence spectrum is about two orders of magnitude lower than that from the ordinary single spectrum and it is found to take a long counting time to get a spectrum with acceptable errors. Therefore, optimizing the coincidence PGNAA setup is critical to improving the coincidence counting rates [5].

2. Simulation tools

A lot of computer simulations shown here were obtained by using the general-purpose Monte Carlo *N*-Particle Transport code MCNP version 5 [6]. However, MCNP5 lacks the capacity to simulate events in coincidence, which is the most important concept of interest here. A specific purpose Monte Carlo code named CEARCPG has been developed at CEAR to generate both the normal and coincidence spectra for the prompt-gamma-ray neutron activation analysis (PGNAA) inverse analysis problem. The algorithm for sampling the neutron-induced prompt gamma

* Corresponding author.
E-mail address: jwang3@ncsu.edu (J. Wang).

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doi:10.1016/j.nima.2010.08.011

rays from the appropriate decay schemes has been developed and implemented within the CEARCPG code, making it possible to calculate the forward coincidence spectrum by the Monte Carlo method. A detector response function (DRF) generation code has also been developed to provide this capability to CEARCPG, which increases the calculation efficiency and/or improves the accuracy by about a factor of 100 [7].

3. System design

3.1. Detector choices

Instead of using two identical NaI detectors, the important part of the optimization is to use one different detector as a "trigger" of coincidence events. This detector will cover as big solid angle as possible to increase the possibility of coincidence detection and has good time resolution. A plastic scintillation detector is a good choice for this purpose because of its lower cost and good absolute efficiency compared with large NaI or BGO crystals. According to the Monte Carlo simulation of MCNP5, a $70 \times 50 \times 10$ cm^3 plastic scintillation detector could provide about 3 times better absolute efficiency than a 6×6 $in.^2$ NaI detector in the energy range 0–10 MeV (Fig. 1). Inherent time resolution of the plastic scintillation detector is also good. Its pulse width is around 2.5 ns with a rising time of around 1 ns. As the main compositions of the plastic scintillation detector are hydrogen and carbon, it has the potential to also be used as a moderator to thermalize neutron source spectra. The drawback of the plastic scintillation detector

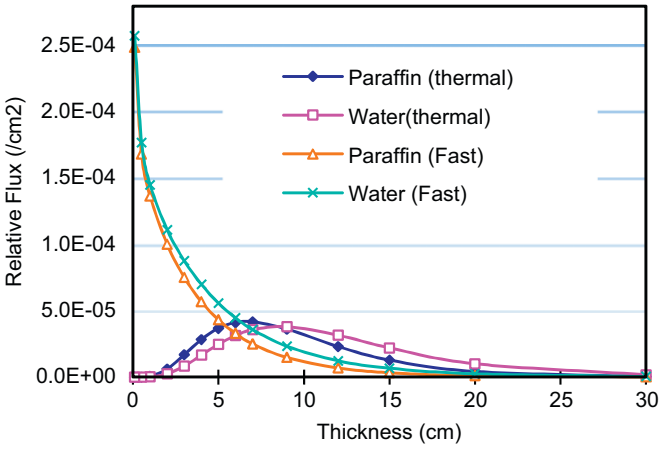

Fig. 3. Moderators comparison between different thicknesses of paraffin and water.

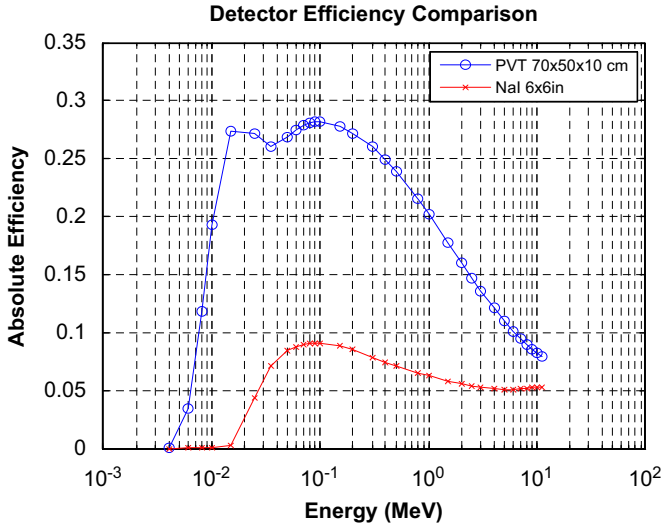

Fig. 1. Absolute detector efficiency comparison between plastic detector and NaI detector (size indicated in figure).

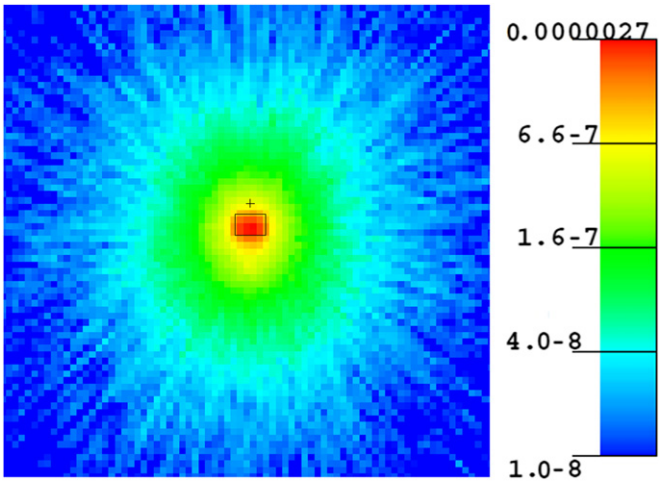

Fig. 4. Photon flux distribution around the small coal sample. Source is facing the center of the bottom face and 10 cm away.

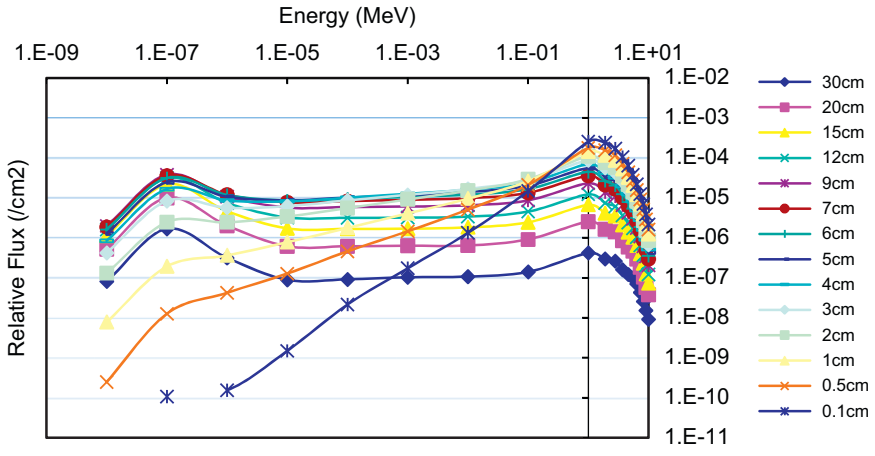

Fig. 2. Energy spectra of ^{252}Cf neutron flux after moderation of different thicknesses of paraffin.

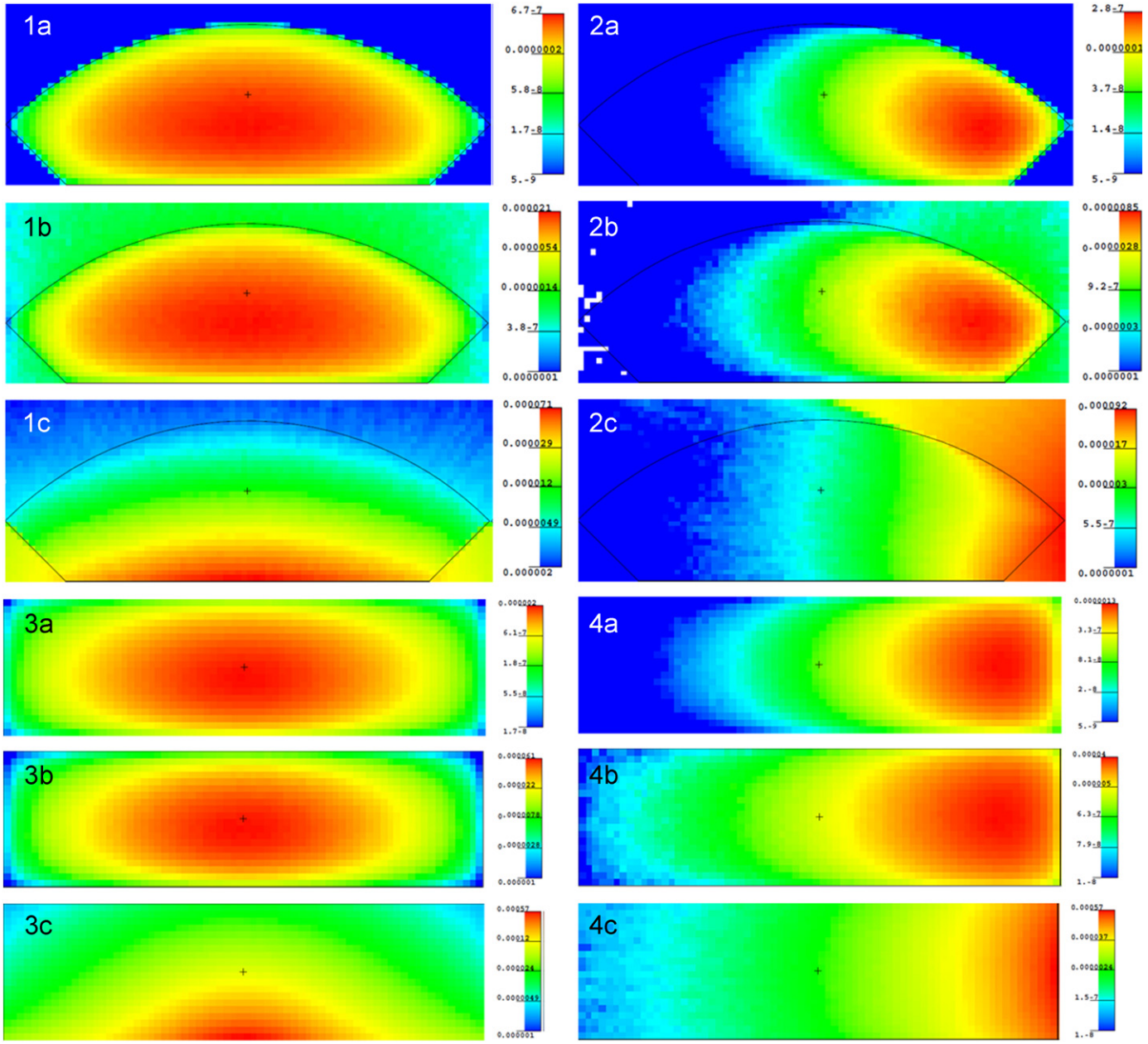

Fig. 5. Neutron flux spatial distribution maps in the large rectangular shape coal sample (bottom two quadrants) and the conveyor belt shape coal sample (top two quadrants). The source is placed under the sample and 30 cm away from the center of bottom surface for the left two quadrants and is placed on one side of the sample and 30 cm away from the center of edge for the right two quadrants. In each quadrant, the top plot is neutron radiative capture rate, the middle plot is thermal neutron (0–0.026 ev), and the bottom plot is fast neutron (1–10 MeV).

is that the main component of its spectrum is the Compton continuum only and it is generally used as a counter, not for spectrum. The reason is that Compton scattering dominates the photon reaction from 20 keV for plastic scintillation detector comparing to 250 keV for NaI detector [8]. However, we can project the 2D coincidence spectrum to the NaI axis to overcome this drawback and utilize its advantages.

3.2. Source moderation

^{252}Cf spontaneous neutron source is one of the most commonly used thermal neutron sources in PGNAA analyzers. It has an average energy of 2.2 MeV, and its neutron energy spectra can be described by the Maxwellian distribution. The energy distribution spectrum of fission gamma rays is from the experimental work by Verbinski et al. [9]:

$$N(E) = Ce^{-E/1.42}E^{0.5} \quad (1)$$

Among all types of neutron interactions, the inelastic scattering interaction and radiative capture interaction are the most important reactions to produce prompt gamma rays. Most of the neutron energy is below the threshold of neutron inelastic reaction for most isotopes in the bulk sample that as of interest, and the neutron capture cross-section is about two order of magnitudes higher than the neutron inelastic reaction and follows

the $1/v$ rule. Thus, thermalizing ^{252}Cf might be needed in certain circumstance, especially for small sample sizes. Self moderation would be enough for large bulk samples as shown later.

Two common moderator materials have been investigated, paraffin and water. As shown in Fig. 2, thermal neutron flux increases first due to thermalization and then decreases due to absorption in the moderator. Through Monte Carlo simulation, 6 cm thick paraffin or 7.5 cm thick water (Fig. 3) has been found to be suitable for a small size sample (9.7 cm × 6.7 cm cross-section in this work). At this thickness, thermal neutrons and fast neutrons are about 50% each, which could have a higher neutron capture (n, gamma) reaction rates and guarantee good penetration into the sample. Due to carbon in the paraffin, the prompt gamma from paraffin will be the interference to the analysis of the bulk sample. Prompt gamma rays from moderator will be less for water than paraffin as oxygen has a smaller cross-section for radiative capture and a higher threshold energy for inelastic scattering.

3.3. Neutron and photon flux distribution

To reach the best coincidence detection, the placement of ^{252}Cf neutron source and two detectors, especially the 6×6 in.^{2} NaI detector, needs to be optimized to detect the maximum amount prompt gamma rays as possible. Thus, the neutron energy flux spatial distribution in the bulk sample and the photon flux spatial distribution around the bulk sample have been simulated to indicate where the prompt gamma rays come from and when they go. Three bulk sample have been investigated, including two rectangular samples (70 cm × 80 cm × 20 cm and 55 cm × 9.7 cm × 6.7 cm) and a conveyor belt shaped sample (45° troughing angle, 60 cm bottom width, 80 cm top width). The bulk material simulated is coal as coal analysis is where PGNAA is widely used.

For the small sample, it is intuitive to place the source facing the center of the biggest surface of the sample. Fig. 4 shows that photons emitted from left–right surfaces are similar to those from top–bottom surfaces, which are both suitable for detection.

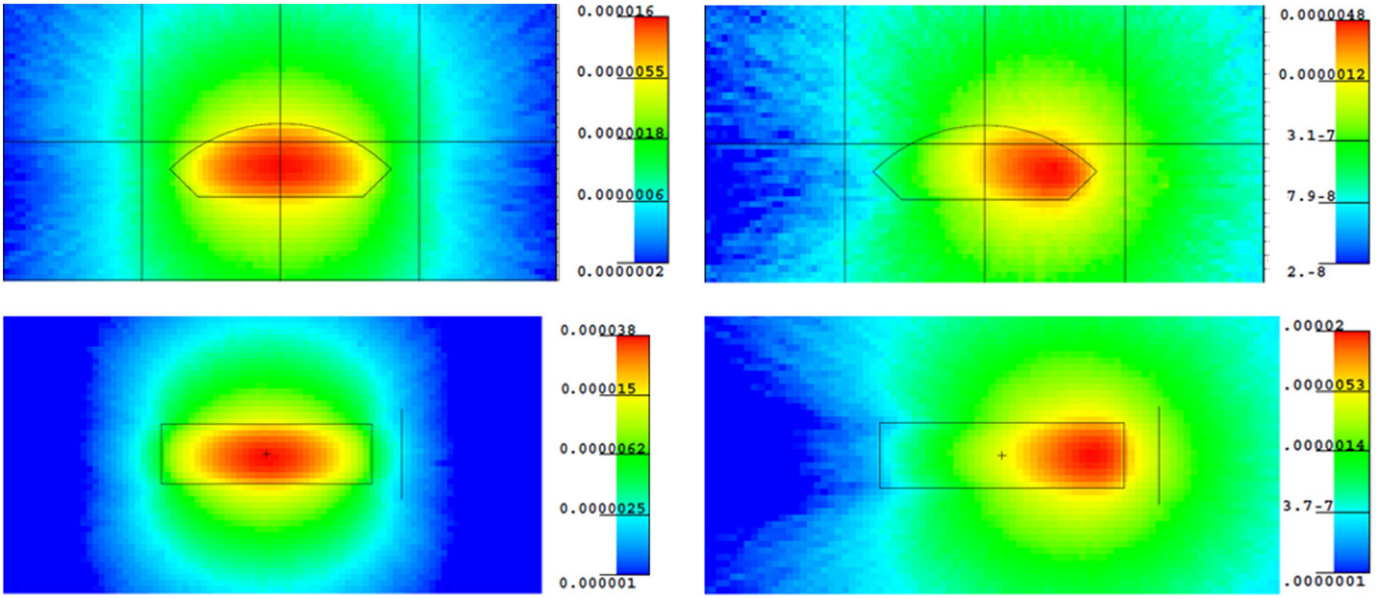

Fig. 6. Photon flux spatial distribution maps around the large rectangular shape coal sample and the conveyor belt shape coal sample. Source placements are the same as described in Fig. 5.

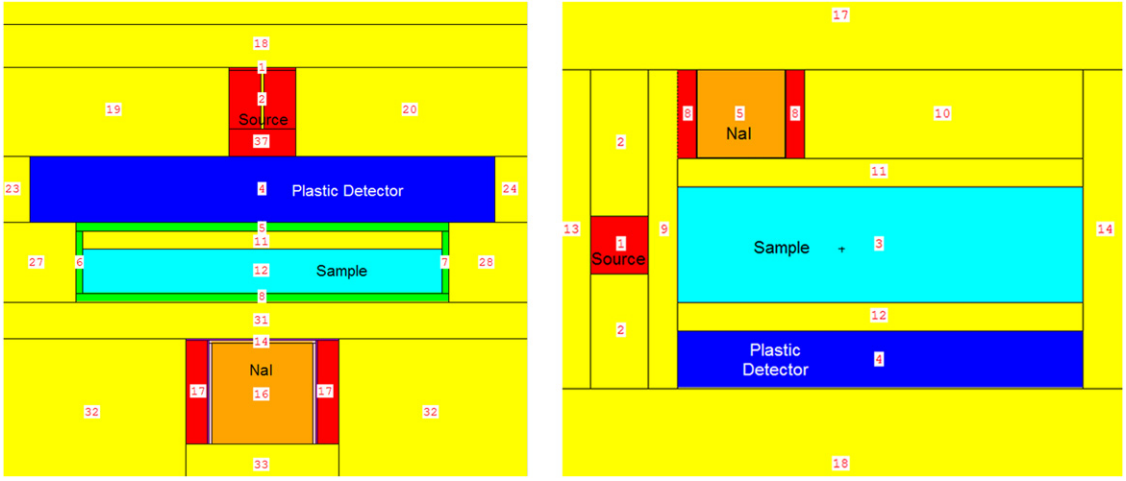

Fig. 7. Two simple preliminary coincidence PGNAA setups.

For the large sample and conveyor belt shape sample, the neutron radiative capture reaction rate maps (Fig. 5-1–4(a)) show where the prompt gamma rays come from, which agree with the thermal neutron (0–0.026 ev) maps (Fig. 5-1–4(b)). These figures show that the radiative capture reactions happens all through the coal sample with the highest production in the center area, if the source is placed under the sample. The reaction is biased to the side near the source with the highest production at 8 and 15 cm distance away from the edge for rectangular and conveyor belt shaped samples separately, if the source is placed at one side of the sample. Fig. 5-1–4(c) displays the fast neutron (1–10 MeV) maps to show the self-moderation of the coal sample. Besides the biased reaction center, the side source position has reaction rates less than 50% of that of the center source position due to the smaller solid angle facing the sample. A more direct indication of the detector placement is in the photon flux maps in Fig. 6, which show a similar conclusion as that from Fig. 5.

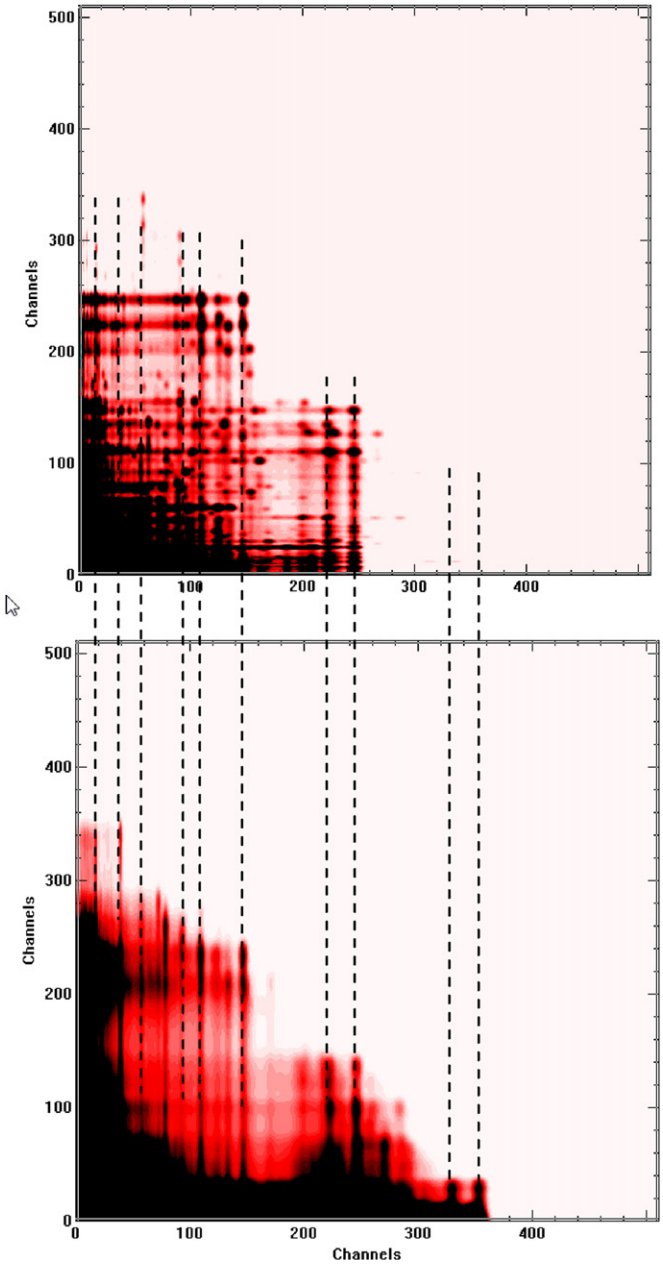

Fig. 8. (Top) 2D coincidence spectrum from two 6×6 in^2 NaI detectors coincidence approach. (Bottom) 2D coincidence spectrum from one $70\times50\times10$ cm^3 plastic detector and another 6×6 in.^{2} NaI detector coincidence approach.

4. Simulation results

Two simple preliminary arrangements have been initialized according to the simulation results shown in (Fig. 7). Through CEARCPG, the 2D coincidence spectrum of the arrangement in Fig. 7 has been simulated. In the left arrangement in Fig. 7, the plastic scintillation detector was also used as neutron moderator between the ^{252}Cf source and the small bulk sample. Overall, the total coincidence signal has been improved by a factor of 3.5 in the simulation.

Fig. 8 shows the simulated 2D coincidence spectra of the old NaI to NaI coincidence arrangement (top) and the new plastic to NaI coincidence arrangement (bottom). As discussed in the previous part, the new coincidence spectrum is not symmetric as the old one was due to the no-peak property of the plastic scintillation detector. However, the axis representing the NaI detector still has good energy resolution. As a benefit of higher counting rates, the two higher energy peaks are easier to observe in the spectrum of the new arrangement.

After projecting the 2D spectrum to the axis representing NaI, we can get the traditional total coincidence spectrum shown in Fig. 9. The simulated sample contains sulfur and a little mercury,

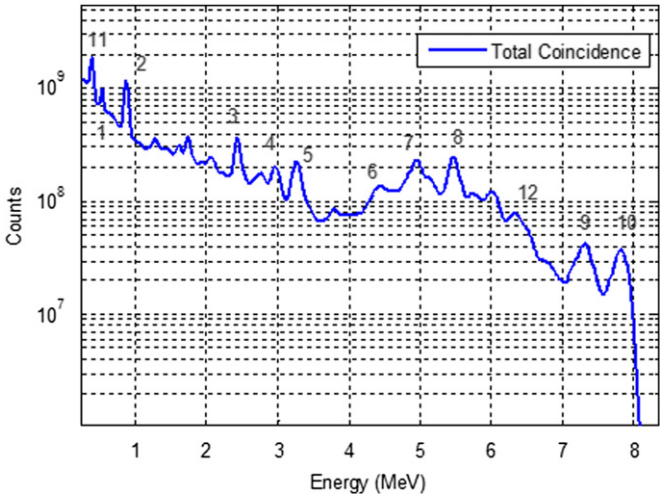

Fig. 9. Total coincidence spectrum after projection to NaI detector axis.

Table 1
Peaks in the total coincidence spectrum from CEARCPG.

Peak	Energy (MeV)	Source
1	0.511	Pair production
2	0.841	Sulfur
3	2.379	Sulfur
4	2.931	Sulfur
5	3.22	Sulfur
6	4.4308	Sulfur
7	4.869	Sulfur
8	5.4205	Sulfur
9	7.31	Hg
10	7.8	Sulfur
11	0.367	Hg
12	6.457	Hg

whose peaks (Table 1) are very clear in Fig. 9. Sulfur and mercury are of great interest in coal analysis.

5. Conclusions and future work

Through MCNP5 simulation, various parameters of the coincidence PGNAA system arrangement have been investigated to find the optimum case to best enable coincidence prompt gamma-ray detection. A preliminary arrangement has also been simulated by the CEARCPG code for the 2D coincidence spectrum between a plastic detector and a NaI detector. The new approach has been shown to work and to be very promising.

A benchmark experiment will be done in the near future to validate the CEARCPG simulation results. To further explore coincidence PGNAA, more 2-D coincidence inverse spectral analysis techniques need to be investigated (like the *Q*-value summing approach) to better utilize the spectral coincidence approach. Effective sample volume of the coincidence approach is also important for determining where the coincidence events emanate from.

Acknowledgements

The authors gratefully acknowledge the support of the CEAR Associates Program on Nuclear Techniques in Oil Well Logging, which includes Baker Atlas, Halliburton, Weatherford, EXXON Mobil, Pathfinder, and Los Alamos National Laboratory.

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