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A feasibility study of a coincidence counting approach for PGNAA applications

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Abstract

Prompt gamma-ray nutron activation analysis (PGNAA) has an inherently low signal-to-noise (S/N) ratio primarily because of the large background (noise) associated with it. Most elements emit a significant fraction of their prompt gamma rays in coincidence with one or more other prompt gamma rays. This paper reports on initial efforts to use coincidence counting in PGNAA to significantly reduce the several sources of background and thereby increase the S/N ratio. An added benefit is the elimination of the often dominant hydrogen prompt gamma-ray spectrum which emits only a single prompt gamma ray with an energy of 2.223 MeV. Preliminary results are given for both in situ bulk analysis applications with a ^{252}Cf neutron source and for nuclear reactor thermal neutron beam applications for small laboratory samples.

Keywords: PGNAA coincidence

1. Introduction

In most prompt gamma-ray neutron activation analysis (PGNAA) applications the inherent signal-to-noise (S/N) ratio is very small. This is caused primarily by the large background (or noise) which has contributions from: (1) natural background sources such as that from ^{40}K, the natural uranium chain, and the natural thorium chain; (2) the gamma rays emitted by the neutron source (either radioisotope or machine sources); (3) the gamma rays produced by neutron interactions from the source within the detector; and (4) cosmic radiation. In addition there are contributions of prompt gamma rays from non-sample materials of construction around or within the PGNAA analyzer and there are sensitivity problems in that certain elements (like hydrogen in particular) yield prompt gamma-ray intensities for typical cases that are one or two orders of magnitude larger than other elements of interest.

It occurred to us that most prompt gamma rays are in coincidence with others and the use of coincidence counting in PGNAA might significantly reduce the background and (possibly) increase the S/N ratio. (Note that the signal is also reduced somewhat by coincidence counting.) In particular the one element that causes problems in many typical cases (hydrogen) is the element that has only one gamma ray at 2.223 MeV with no coincidences. It would appear that coincidence counting in PGNAA would eliminate all the natural background sources that have no coincidences,

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the gamma rays emitted by the neutron source (unless they were in coincidence with other gamma rays such as the prompt gamma rays produced in surrounding materials), the gamma rays produced in individual detectors by source neutron interactions, and that from cosmic radiation. Coincidence counting greatly favors response from radiation sources that are close to both detectors. This means that coincidences produced in surrounding materials (not the sample) would be depressed relative to those produced close by, as in the sample. So some sample volume would be sacrificed for this decrease in background. Note that the removal of the very intense hydrogen spectrum opens up the possibility of using many low-energy prompt gamma rays that are very abundant in many important elements such as those in sulfur (0.841 MeV with a 75.6% abundance) and carbon (1.262 MeV with a 29.5% abundance).

Unfortunately, most coincidence schemes for the elements are not presently known. The schemes for a few of the low atomic number elements (like carbon and oxygen) are obvious, but not those with a larger number of prompt gamma rays. These schemes can be ascertained by using prompt gamma-ray abundances with the 'Q_n' values of the reaction with added help from the nuclear levels (Firestone, 1996). The existing prompt gamma-ray abundances (Lone et al., 1981) are generally not accurate enough to preclude coincidence scheme ambiguities. There is a need for computer trial and error searches and experimentation to determine or verify most of the elemental (and isotopic) coincidence schemes.

2. Preliminary feasibility studies

The present feasibility study has both an experimental part and a simulation part. In the initial experimental part, the experimental arrangement used a 5 μg ^{252}Cf source for neutrons and two 5 in. × 5 in. NaI crystals in the coincidence counting mode. Many of the prompt gamma-ray peaks of sulfur were clearly detected (especially the most abundant ones) and a few peaks of other sample elements were also detected. When this partial success was achieved, another feasibility study was performed using the PULSTAR thermal neutron beam facility as the neutron source. The problem of high background gamma rays present in the PULSTAR reactor neutron beam adversely affected the sensitivity of the system and was the primary source of random coincidences that dominated the spectra that were taken. Then, believing that the thermal neutron beam at the National Institute of Standards and Technology (NIST) (Lindstrom et al., 1995) would be much cleaner (the beam is far from the reactor and is well-collimated), another feasibility study was performed there. Primarily due to the neutron scattering of a neutron lens at the same measurement location, the NaI crystals were activated and this minimized the system sensitivity. Currently, shielding of the two detectors against thermal neutrons using a ^{6}Li sheet and collimating the detectors with lead cones is in progress for another feasibility study at NIST. It is also planned to re-visit the use of the ^{252}Cf source for bulk sample applications.

In the simulation part, the PULSTAR reactor experiment was simulated using Monte Carlo techniques. Four different experimental arrangements for the relative positions of the two detectors with respect to the neutron beam and the sample orientation were investigated.

3. Coincidence scheme concepts

Fig. 1a illustrates the concept of coincidence measurements. In this very simple single channel system, the basic technique is to convert the analog signal from the detectors into a logic signal and then send these pulses to a coincidence module. If the two signals are "coincident", then a logic signal is produced at the output; if not, no signal is produced. Thus all pulses arriving within a time equal to the sum of their widths, called the "coincidence time", are recorded as "coincident". Fig. 1b shows some examples of coincident and non-coincident pulses.

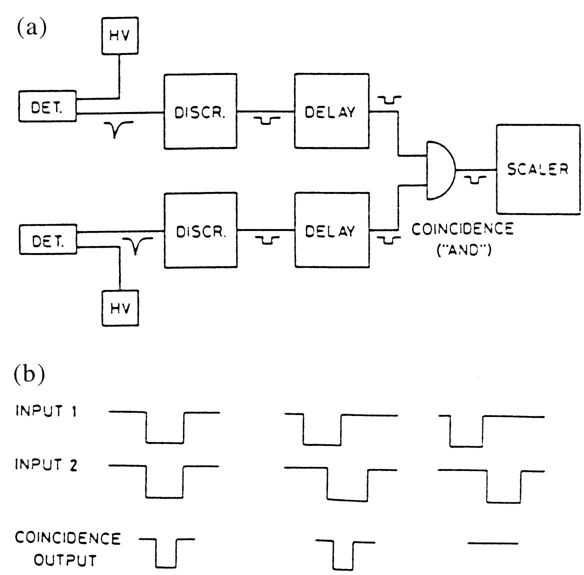

Fig. 1. (a) A simple coincidence circuit.(b) Expected coincidence output.

In addition to the coincident events, each detector will produce pulses that correspond to "chance coincidences". Sources of chance coincidences may be the coincidence between any of the prompt gamma rays and any kind of background mentioned earlier. Another source of background is from "cross-talk" between the two detectors where a gamma ray enters one detector, deposits part of its energy, and then escapes and enters the other and deposits part or all of its remaining energy. This kind of background is minimized by shielding the two detectors from one another.

The intensity of chance coincidences depends primarily on the counting rates in both detectors and on the coincidence time according to:

$$R_C = \tau N_1 N_2 \tag{1}$$

where R_C is the chance coincidence counting rate, τ is the coincidence time, or roughly speaking the resolving time, and N_1 and N_2 are the counting rates of each detector, respectively. One would normally like to minimize the chance coincidence counting rate. This can be accomplished by minimizing the

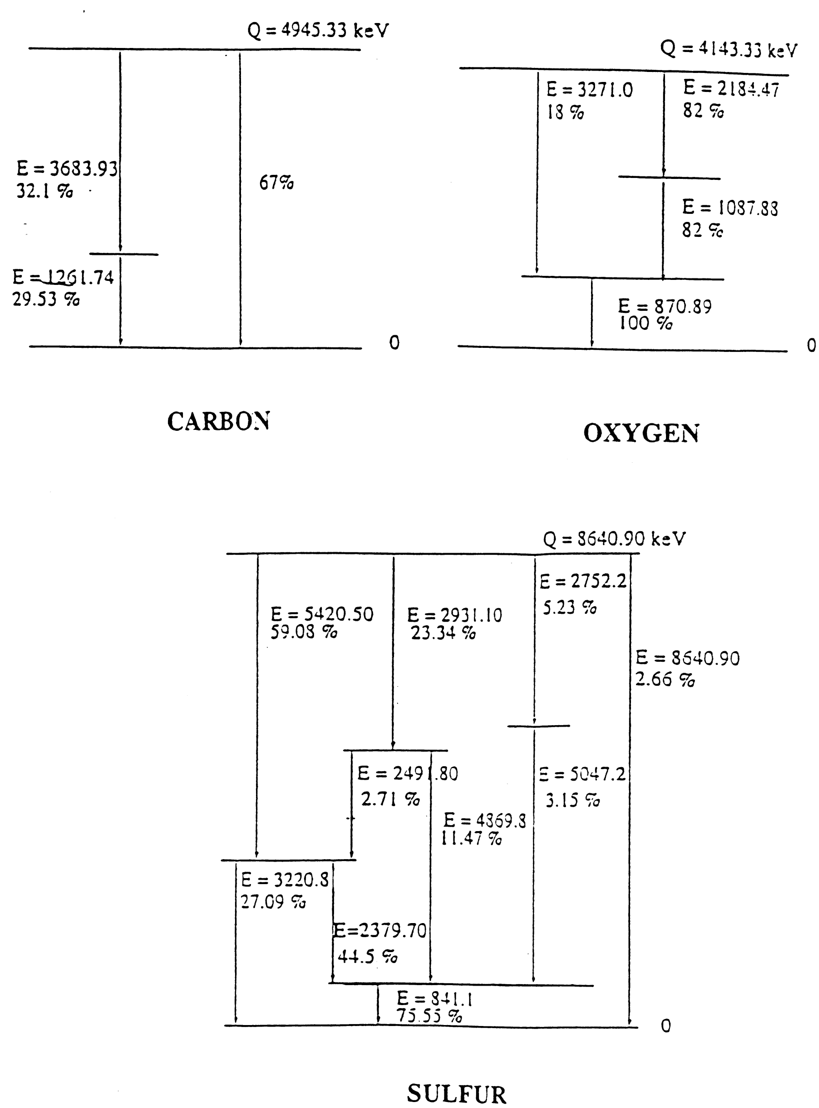

Fig. 2. Proposed coincidence schemes for some light elements of interest to PGNAA.

resolving time in the coincidence unit and/or making use of an energy selection criterion that can limit the counting rates from each detector by discarding any events that have a low or zero probability of yielding true coincidences.

Fig. 2 shows schematic diagrams of the proposed coincidence schemes of some of the light elements of interest to the best that they could be predicted using the existing compilation (Lone et al., 1981) of prompt gamma rays from thermal neutron capture. As the number of prompt gamma rays emitted from the element gets larger, prediction of the coincidence schemes become more difficult. Currently, attempts to refine the predicted coincidence schemes of the elements of interest are in progress, along with the prediction of the schemes of other important elements.

4. Advantages of the coincidence approach

There are two important advantages of using coincidence techniques in PGNAA. The first is the elimination of most of the interfering background sources. This includes: (1) the hydrogen spectrum, which is often orders of magnitude higher than that for other elements and interferes with many prominent prompt gamma rays from most of the elements of interest, (2) background produced by thermal neutron activation of both Na and I within the NaI crystal when NaI detectors are used, (3) background gamma-rays from the analyzer structure materials and other surrounding structures, (4) natural background (^{40}K, Uranium, and Thorium series), and (5) the gamma-ray background from the neutron source.

The second advantage of coincidence techniques in PGNAA stems from the fact that prompt gamma-ray spectra from individual elements typically consist of about 100 gamma rays (see Fig. 3 with data from Lone et al., 1981) with photon energies from almost zero up to about 11.5 MeV. A sample material consisting of a few elements can generate gamma-ray spectra with several hundred individual lines while a complex matrix can generate spectra with several thousand. With such spectra, even high resolution detectors must contend with overlapping peaks. The commonality of many nuclear levels also leads to an even higher overlap potential. Since the use of coincidence techniques should identify the gamma rays emitted at the same time from the same element ("true coincidences"), this should simplify spectra and make elements easier to identify and quantify.

Unfortunately, the use of coincidence techniques is expected to reduce the output signal somewhat and also complicates the detection system. The hope is that the S/N improvement from background (noise) reduction more than offsets the degradation of the S/N by signal reduction.

To improve the sensitivity of the coincidence system, one can impose an energy selection criterion, i.e. energy window, on the triggering detector. Pulses within that window are the only pulses to trigger the gating unit. This idea can be used to look for the coincidence schemes of different elements by setting a window around one of the peaks and observing the resulting coincidence spectrum. The resulting spectra should consist of just those gamma rays that are in coincidence with the triggering gamma ray. At least two windowing options are possible:

(1) equal-size triggering windows at different energies and
(2) variable-size triggering windows around the same energy.

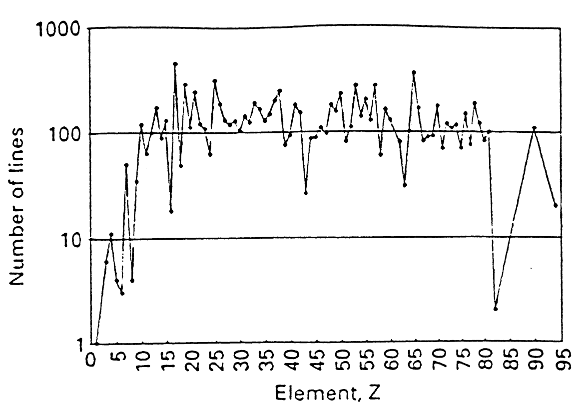

Fig. 3. Thermal prompt gamma-ray lines per element.

5. Monte Carlo simulation

The reactor experiment was simulated using Monte Carlo techniques. The simulation was accomplished as

Table 1
Summary of Monte Carlo simulation results

Arrangement	Carbon (counts/s)	Oxygen (counts/s)	Sulfur (counts/s)
1	197.05 ± 0.20	31.989 ± 0.019	17320.3 ± 10.3
2	9.347 ± 1.853	1.744 ± 0.00225	971.22 ± 1.27
3	9.094 ± 0.0178	1.677 ± 0.0021	949.33 ± 1.23
4	48.456 ± 0.060	8.773 ± 0.0066	4867.47 ± 3.75

(a)

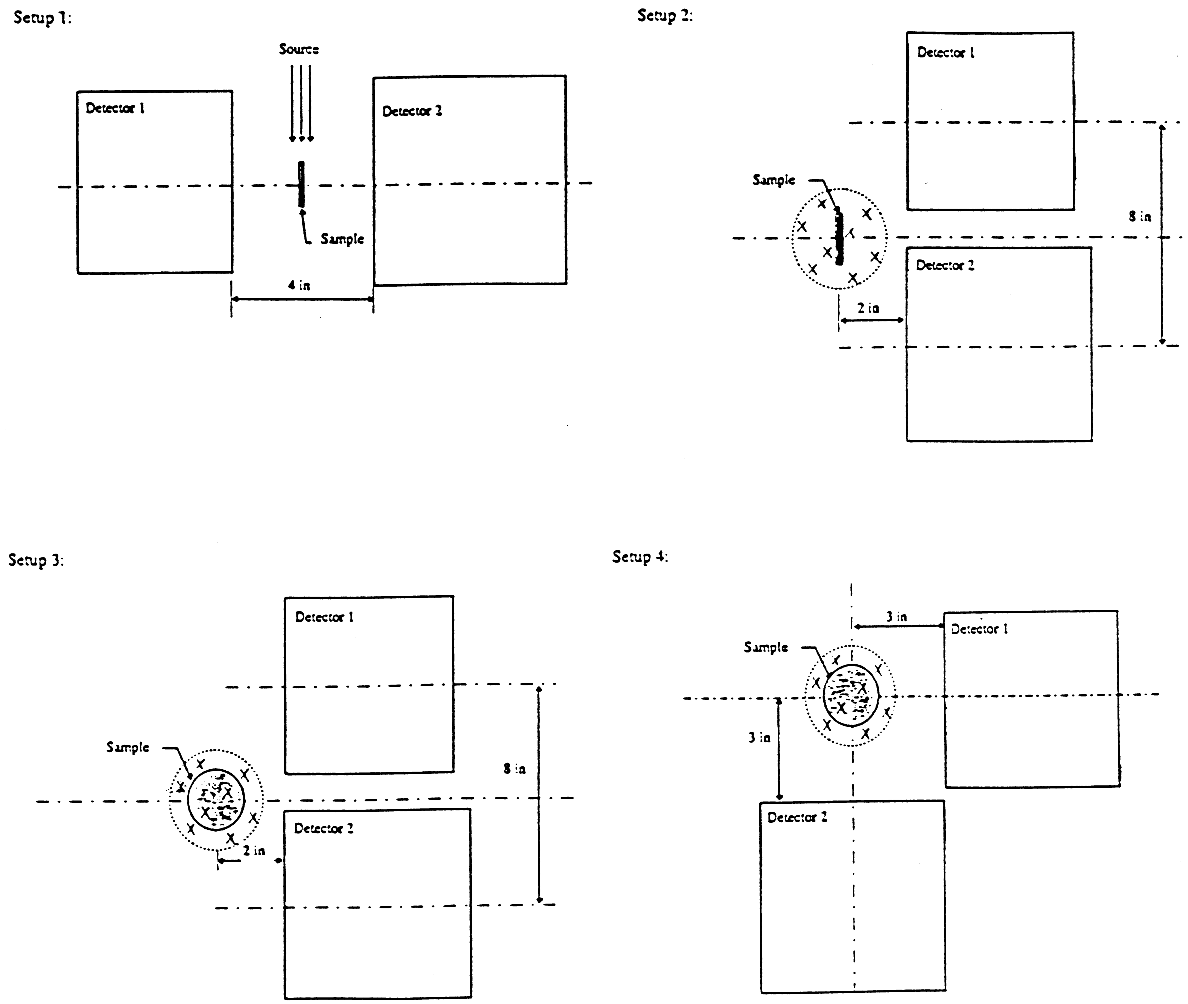

Fig. 4. (a) Different experimental arrangements simulated by Monte Carlo.

follows. A thermal neutron beam with a diameter larger than the sample size is assumed. A capture reaction is forced to occur within the sample volume. At the site of interaction, one of the coincidence schemes is chosen based on its relative abundance and all the prompt gamma rays in that scheme are emitted (at present assumed to be isotropic). Each one of the emitted coincident prompt gamma rays is tracked separately and the detailed energy deposition in each of the two detectors is banked. After tracking all the coincident prompt gamma rays of the chosen scheme, the energy deposited in the triggering detector is examined; if it is greater than zero, the response of the other detector is recorded in the proper channel of the spectrometer; if the energy deposited in the triggering detector is still equal to zero, the response of the other detector is ignored.

Four different arrangements for the relative positions of the two detectors, the neutron beam and the sample orientation were investigated. Fig. 4a shows the four arrangements investigated and Table 1 summarizes the results. Fig. 4b shows the predicted coincidence spectrum of sulfur. Results showed that the highest counting rate is obtained when the two detectors are facing each other. When the cross-talk component was tallied separately, it turned out that it is a

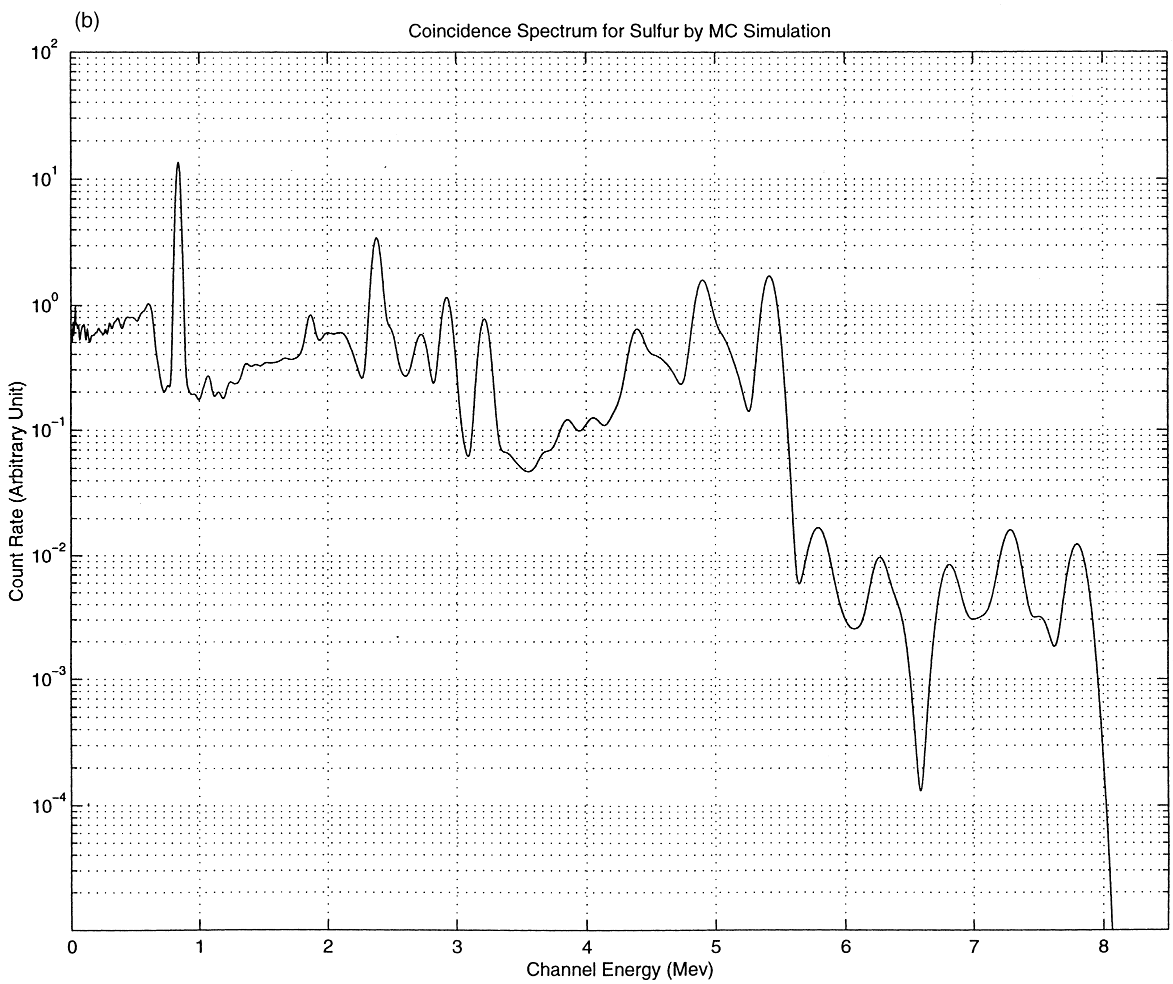

Fig. 4. (b) Sample Monte Carlo results for sulfur.

major contributor in the total counting rate for this arrangement. The optimum arrangement is when the two detectors are at right angles to each other because this gives a relatively higher counting rate with a reduced cross-talk component. Currently, more simulation of the possible coincidence arrangements is in progress.

6. Experimental work

The first feasibility study was performed in the laboratory using a ^{252}Cf source as a neutron source. The experimental arrangement is shown in Fig. 5a and the electronics used are shown in Fig. 5b. When a capture reaction occurs in the sample box, prompt gamma rays are emitted. If one of the emitted prompt gamma rays is detected by the triggering detector (a 5 in. × 5 in. NaI Crystal), an ENABLE signal for the linear gate is generated. If another pulse comes to the other detector within the resolving time of the circuit, it is considered to be in coincidence with the triggering one and is recorded in the output spectrum.

The neutron source was a 5 μg (on 8/20/91) ^{252}Cf source. The source holder was a 4 in. cube of pure lead for shielding against source gamma rays, followed by an effective vertical thickness of pure paraffin of 7.75 in. to thermalize the fast neutron component. A large sample box (21 in. × 8.5 in. × 4 in.) was used because a small disk sample tried at first did not provide sufficient sensitivity. Finally, because cross-talk between the two detectors is thought to be a major factor in the measurement sensitivity, a 1 in. lead wrapping around the two detectors was used.

The resolving time of the electronic circuit was improved by passing the output of each of the two detectors through an SCA which emits a 500 ns square pulse, then the two output square pulses of the two SCAs were fed into a coincidence module modified by introducing a shunt capacitor at its input to differen-

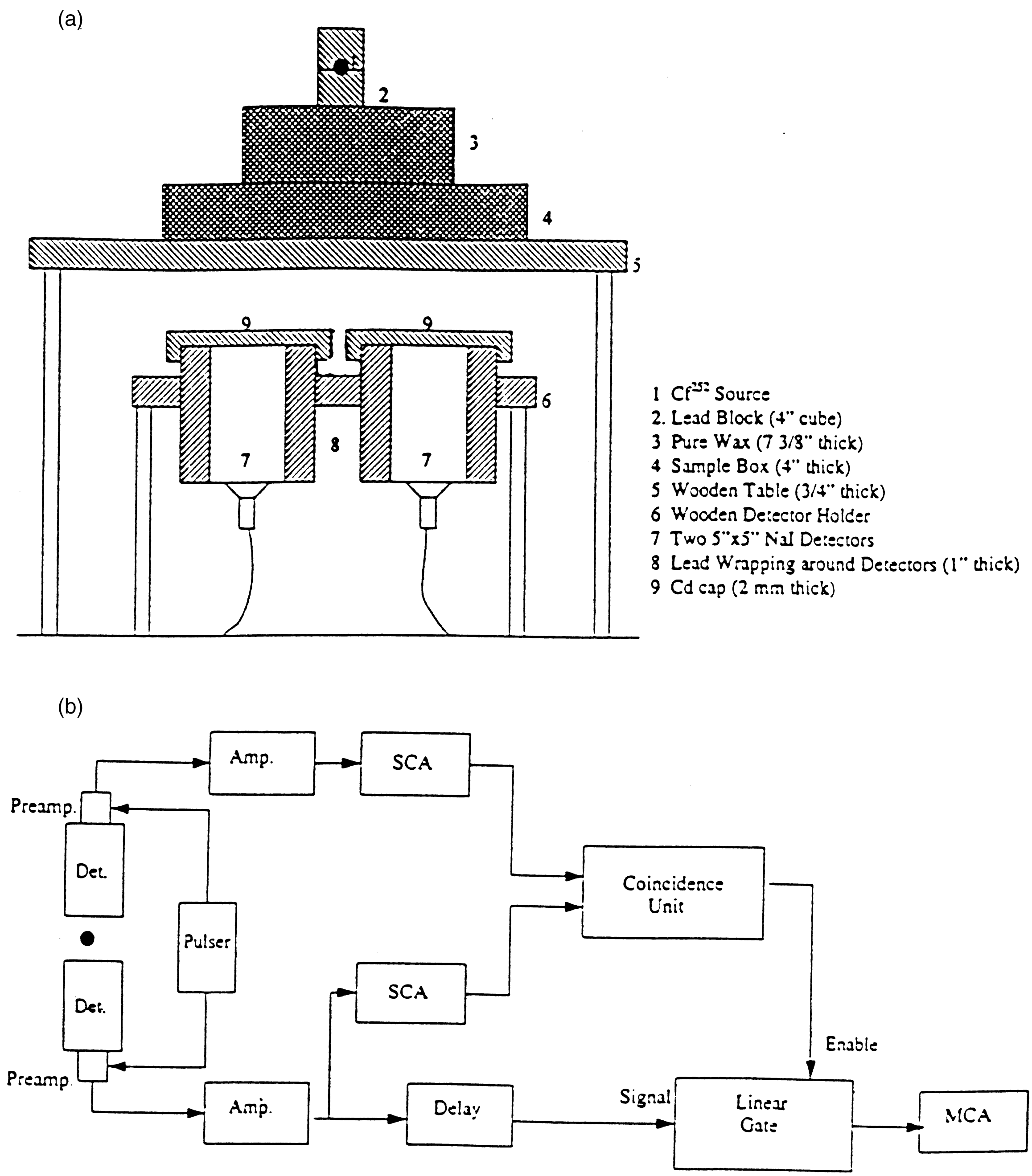

Fig. 5. (a) Refined arrangement of the lab experiment. (b) Refined electronics circuit for both the lab and reactor experiments.

tiate the input signal and reduce the resolving time to almost one-tenth of the original value. The resolving time of this circuit was measured using a 60 Hz pulser and was found to be 80 ± 25 ns.

The sample elements were chosen based on their importance to PGNAA applications. They include: sulfur in the form of pure powder, oxygen in the form of water because hydrogen in the water has no coincidences and there is no problem with its interference, nitrogen in the form of guanidine carbonate ($C_2H_{10}N_6H_2CO_3$) which was chosen because it has a higher ratio of nitrogen to other elements in the compound compared to other available nitrogen-bearing materials, carbon in the form of pure graphite blocks, and chemically stable chlorine in the form of NaCl.

The first material examined was sulfur since it has a high capture cross-section and is of major interest in coal analysis. Fig. 6 shows a sulfur spectrum for both a single detector system and a coincidence system. Most of the high abundance prompt gamma rays of sulfur can be identified in the coincidence spectrum while some of them are hidden in the single detector response spectrum under other components (annihilation radiation, hydrogen peak, natural background, NaI activation, etc). The big advantage of the coincidence system in eliminating the dominant hydrogen peak is evident in this case. The most abundant prompt gamma ray for sulfur at 0.841 MeV is very clear in the coincidence spectrum. Elements which have lower cross-sections than sulfur and/or have a more flat prompt gamma ray distribution did not give results as good as those for sulfur. Examples are nitrogen, as shown in Fig. 7, and oxygen, as shown in Fig. 8. Elements like carbon and chlorine

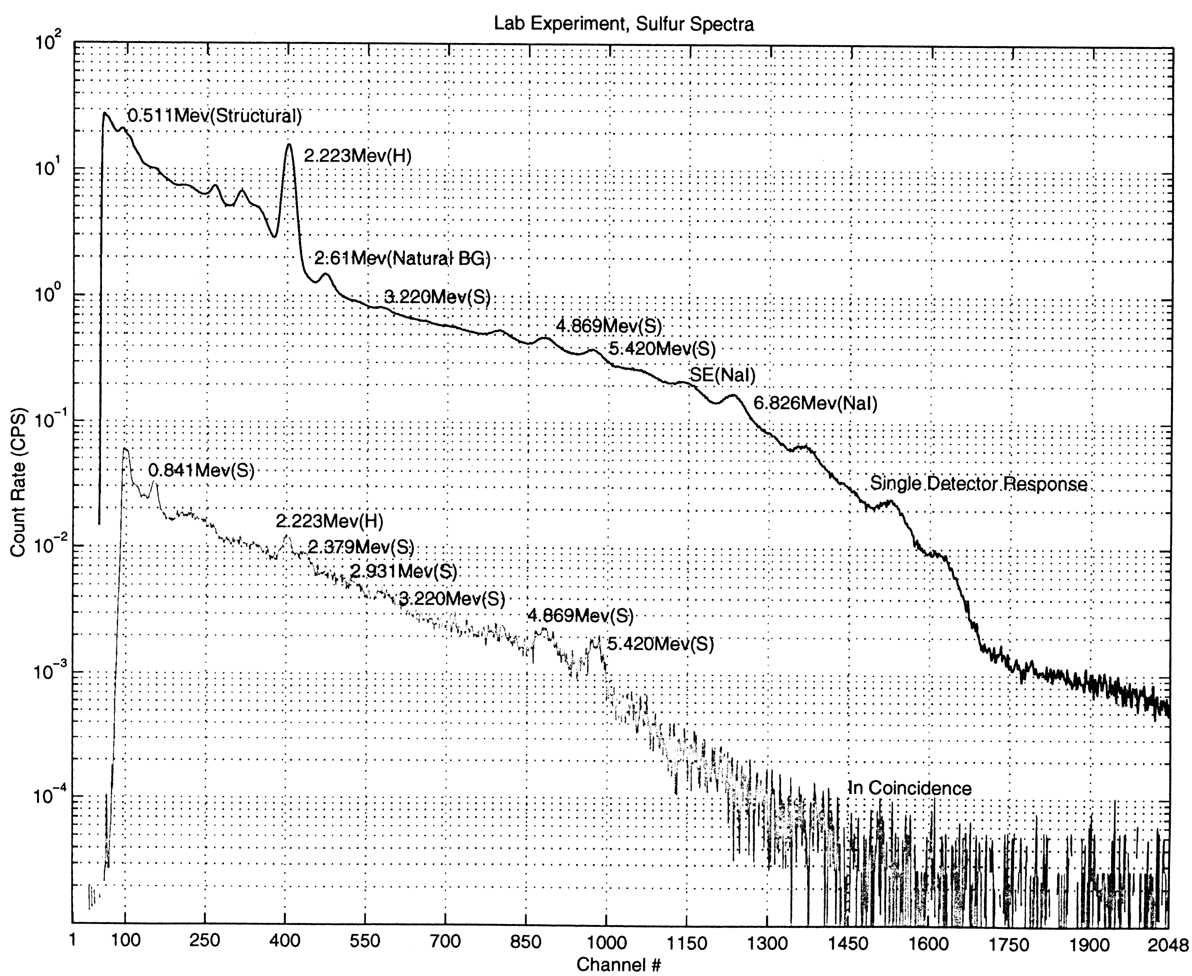

Fig. 6. Lab experiment: sulfur spectra; single detector and coincidence responses.

did not show any sensitivity with the existing coincidence system.

The ^{252}Cf experiment indicated that the idea is feasible. We decided to move on to experiments on the thermal neutron beam of the NCSU PULSTAR reactor. This should give a better thermalized neutron source, a higher neutron intensity, and a very localized neutron beam. All of this should help in reducing the sample size which, in turn, should reduce the self-attenuation effect of the emitted prompt gamma rays. The same electronics circuit was used. Experiments were performed over a variety of power levels from 1W to 100 kW. Sensitivity was found to be worse than that of the lab experiment. This can be attributed to the high, varying gamma ray background that accompanies the neutron beam. Also, a fast neutron component in the neutron beam is thought to exist. The whole experimental arrangement was placed inside a helium-filled bag to reduce scattering, but no significant improvement was observed.

To study how much contribution to the resulting spectrum is coming from random coincidences, Eq. (1) was implemented channel by channel and the results are shown in Fig. 9. It is obvious that random coincidences dominate the resulting spectrum.

The PULSTAR neutron beam experiment has shown the importance of the detectors being placed in a clean field (clean from source neutrons, source gamma rays, and scattered gamma rays). At NIST, the neutron beam is far from the reactor. A detailed description of the NIST facility can be found in the article by Lindstrom et al. (1995). The detectors were set up at a 30° angle as shown in Fig. 10, primarily because of work space requirements. Unfortunately,

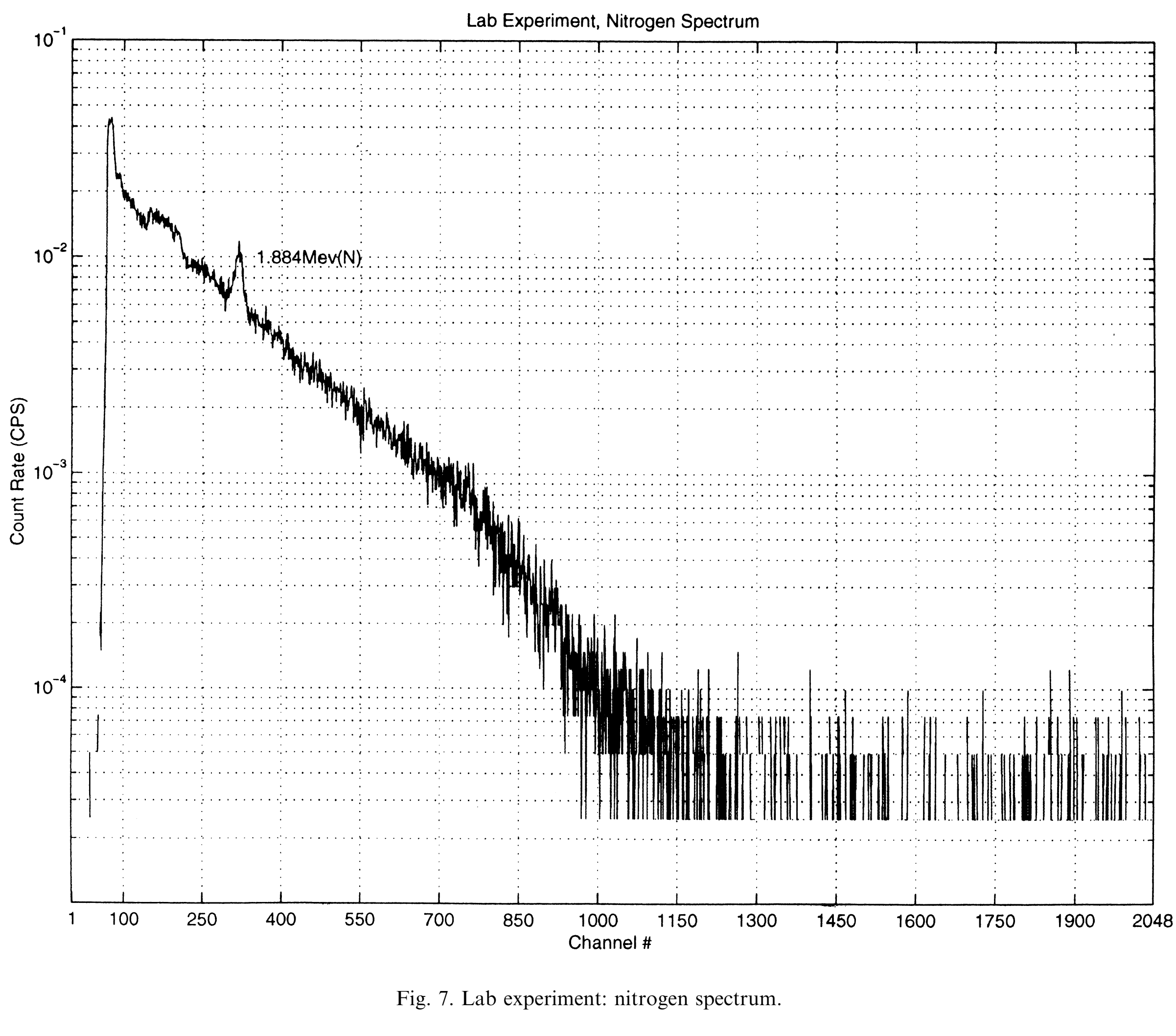

Fig. 7. Lab experiment: nitrogen spectrum.

after the NIST beam was turned on, we realized that there was a neutron lens installed in the immediate vicinity which scatters most neutrons (90%) over the surrounding space. Thus the detectors were neutron activated because there was one side of each detector that was not covered by a ^{6}Li sheet. Currently, a ^{6}Li shield is being designed to shield the detectors against thermal neutrons and the experiment will be performed again.

It should be mentioned that samples for the NIST experiments are very small disks of the material of interest. With good detector shielding and a faster coincidence system, the desired results should be obtained. The following samples are ready for use at NIST: C (graphite), Mn, Mg, NaCl, S, NaI, KI, N (Urea), MgO and P.

7. Discussion and future work

The Monte Carlo simulation feasibility studies undertaken to date have concentrated on the "signal" coincidence counting rates. Future work in this area will include sources of background which will allow realistic estimates of practicality to be evaluated. These sources of background will include random coincidence counting rates and "cross-talk" coincidences that occur when a gamma ray interacts with both of the detectors at the same time. This cross-talk might occur when a pair production or Compton scatter event occurs in the first detector and an escaping photon (annihilation photon or Compton scatter photon) interacts with the other detector.

The present experimental feasibility investigations

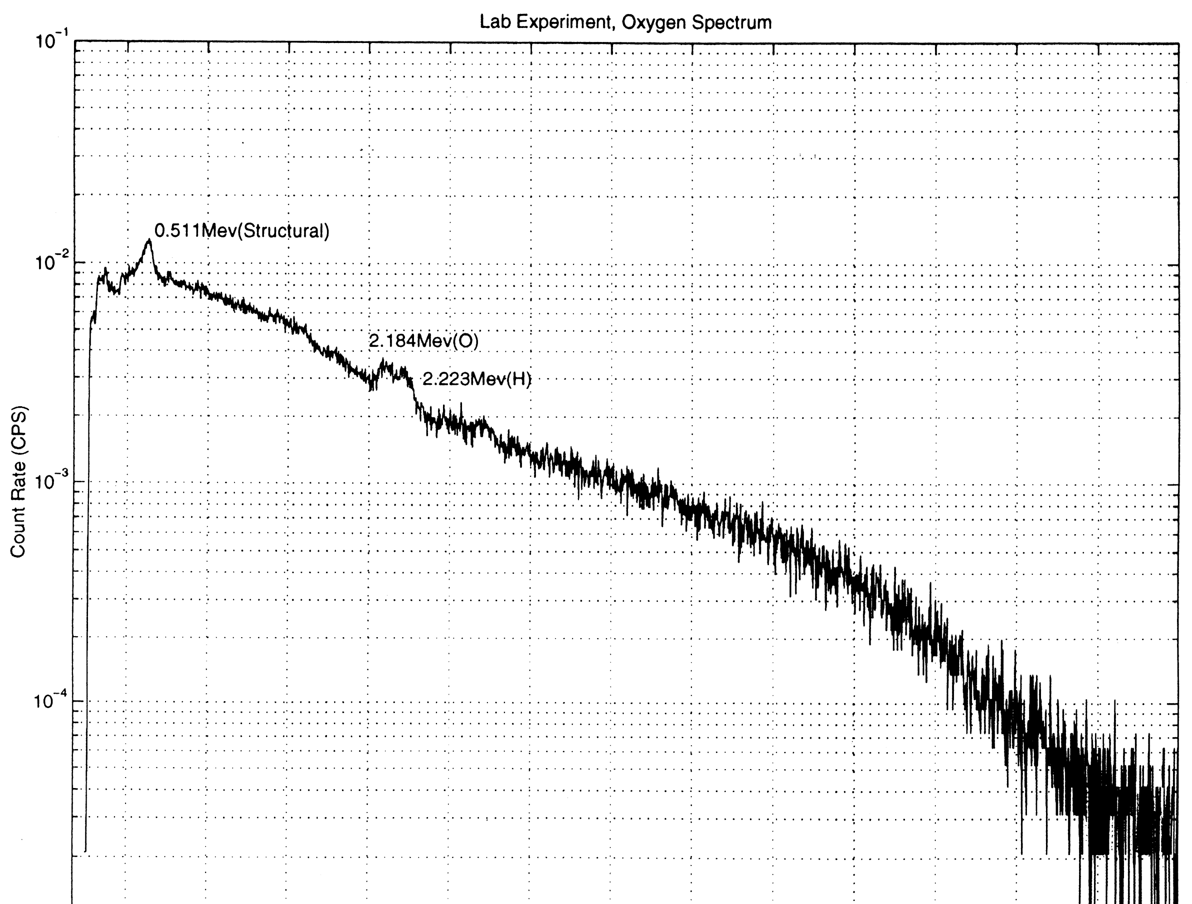

Fig. 8. Lab experiment: oxygen spectrum.

indicate that the coincidence approach may be promising for PGNAA applications. The primary problem with the experimental investigations so far have been in the unexpected high background or noise levels that have been observed. This has come about for probably two main reasons that are exhibited in different degrees in the different experimental arrangements. These are: (1) the random coincidence counting rate from external gamma-ray counting rates incident on the two individual detectors and (2) the random coincidence counting rate from internal gamma-ray counting rates produced by neutron activation of the detectors. The bulk experiments with a ^{252}Cf neutron source were probably dominated by the first, the NCSU PULSTAR nuclear reactor thermal neutron beam experiments had about equal components of both, and the NIST thermal neutron beam experiments were dominated by the second.

With this in mind future work is going on in parallel along the following lines:

(1) ^{6}Li shielding of both detectors against thermal neutrons,
(2) obtaining and using a faster coincidence circuit,
(3) re-visiting ^{252}Cf as a neutron source,
(4) prediction of more accurate coincidence schemes by implementing the energy selection criterion (energy windowing),
(5) use of samples of higher purity (NIST samples),
(6) use of semi-conductor detectors instead of NaI detectors (or a combination of both where the NaI

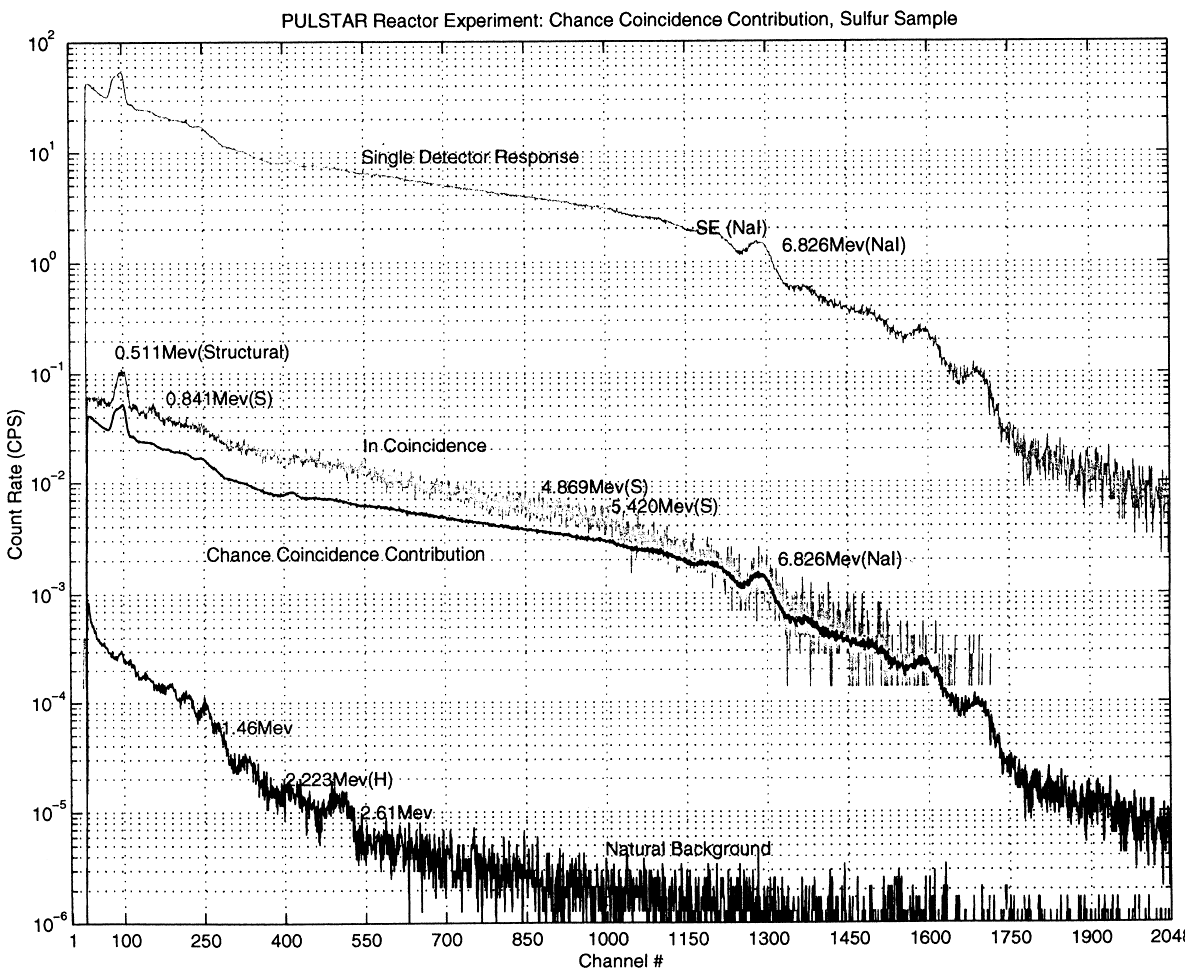

Fig. 9. PULSTAR experiment: sulfur spectra; contribution of chance coincidences.

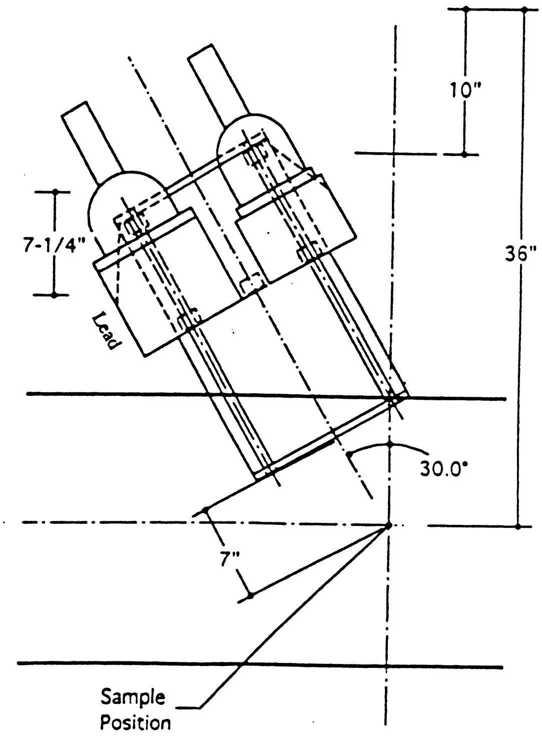

Fig. 10. Detector arrangement in the NIST experiment.

detector triggers and the semi-conductor detector records), and

(7) more concerted efforts devoted to the Monte Carlo simulation of the problem to give a better prior understanding of optimum experimental arrangements.

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