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## SENSITIVITY OF LDEF FOIL ANALYSES USING ULTRA-LOW BACKGROUND GERMANIUM VS. LARGE NaI(Tl) MULTIDIMENSIONAL SPECTROMETERS

James H. Reeves  
Richard J. Arthur  
Ronald L. Brodzinski  
Pacific Northwest Laboratory\*  
Richland, Washington 99352  
Phone: 509/376-2135, Fax: 509/376-2329

### ABSTRACT

Cobalt foils and stainless steel samples were analyzed for induced  $^{60}\text{Co}$  activity with both an ultra-low background germanium gamma-ray spectrometer and with a large NaI(Tl) multidimensional spectrometer, both of which use electronic anticoincidence shielding to reduce background counts resulting from cosmic rays. Aluminum samples were analyzed for  $^{22}\text{Na}$ . The results, in addition to the relative sensitivities and precisions afforded by the two methods, are presented.

### INTRODUCTION

High-purity germanium (HPGe) diodes with photopeak efficiencies greater than 100% relative to a standard 3 in.  $\times$  3 in. cylindrical NaI(Tl) scintillation crystal are now available. HPGe gamma-ray spectrometers have become the instrument of choice for low-background counting in most laboratories with a gamma-ray counting facility because of their high energy resolution and the large sizes currently available. However, there are instances where large NaI(Tl) detector systems offer better results. The measurement of  $^{22}\text{Na}$  in random shaped samples of aluminum of various thickness retrieved from the Long-Duration Exposure Facility (LDEF) is an example. Measurement of  $^{60}\text{Co}$  in many stainless steel and cobalt foils can also be made with more precision and accuracy by counting between two large NaI(Tl) detectors operating in coincidence with each other and in anticoincidence with an active veto shield. For radionuclides in which the gamma rays are in coincidence and have at least moderate energies, NaI(Tl) systems are competitive with even the largest and lowest-background germanium detectors in effective resolution, Compton interference, system background, and counting efficiency. In addition, large dual-crystal NaI(Tl) systems vastly reduce geometrical uncertainties when compared to results obtained using single-diode HPGe detectors. Such NaI(Tl) systems have been previously described in the literature (ref. 1).

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## MEASUREMENTS: HPGe AND NaI(Tl) SPECTROMETERS

An ultra-low background HPGe gamma-ray spectrometer as illustrated in Figure 1 was used for these measurements (refs. 2-3). The system is located above ground in our laboratory and has the lowest background of any above-ground device known to be operating. The NaI(Tl) detector system, located in the same counting room, is pictured in Figure 2. Comparisons of counting efficiencies and system backgrounds for  $^{60}\text{Co}$  are given in Table 1 along with those for a low background germanium system located 50 ft. underground at the Savannah River Laboratory in Aiken, SC, which uses a 90% HPGe crystal (ref. 4). Counting efficiencies and backgrounds use the summation of the peak areas for the 1172.7- and 1332.5-keV gamma rays.

The 31.5% HPGe detector was efficiency calibrated for  $^{60}\text{Co}$  with a 5.08-cm square source with thickness of approximately 0.03-mm supplied through Alan Harmon, Marshall Space Flight Center. The thin source was positioned on the detector side of a sample (or sample blank) for one counting interval and positioned on the opposite side for a second counting interval. The efficiency was determined from the average of the two counts. Efficiencies and backgrounds for the 90% HPGe detector were derived from data contained in ref. 4.

The NaI(Tl) spectrometer was efficiency calibrated using 9.53-cm diameter right circular cylinders containing various thicknesses of sea sand and/or copper powder homogeneously mixed with known amounts of NIST-traceable radionuclides. Tests conducted using copper powder vs. sea sand of thicknesses up to 5 g/cm<sup>2</sup> show differences of less than 1%. Gamma-ray attenuation in the sample material is primarily due to Compton interactions which are not dependent on atomic number (Z).

Three cobalt samples and 5 stainless steel samples were analyzed for  $^{60}\text{Co}$  with the NaI(Tl) system. All were about 5 cm square but varied in thickness. The three cobalt samples and one of the stainless samples were also analyzed with the HPGe spectrometer. Counting intervals were about one week in all cases. The results are shown in Table 2.

The activities determined for the cobalt samples I-C9 and G12-A-1-F-N with the NaI(Tl) spectrometer are higher than the activities from the HPGe system while the activity determined for sample I-H12 with the NaI(Tl) spectrometer is comparable to the activity determined with the HPGe system. Sample I-H12 consisted of two components about 1.91-cm wide by 5.08-cm long and was analyzed as one sample of dimensions 3.82 cm by 5.08 cm. The placement of activity on the 5-cm-square thin-film standard SC-12-Pt90 may approximate sample I-H12 more closely than samples I-C9 and G12-A-1-F-N. There are no corrections made for sample inhomogeneity or for non-uniform activity within the standard. Additionally, the NaI(Tl) system is known to be less sensitive to location of activity within a sample than is a single HPGe detector.

Measurements were performed to establish the uncertainties introduced to the data resulting from geometrical variations within the confines of a 1.27-cm thick by 8.255-cm square geometry. A 9.5-mm diameter  $^{22}\text{Na}$  source was used on both the NaI(Tl) spectrometer and the HPGe detector since the triple coincidence afforded by  $^{22}\text{Na}$  (the annihilation photons plus the 1274.5-keV gamma ray) offers a more stringent requirement than a simple coincidence such as offered

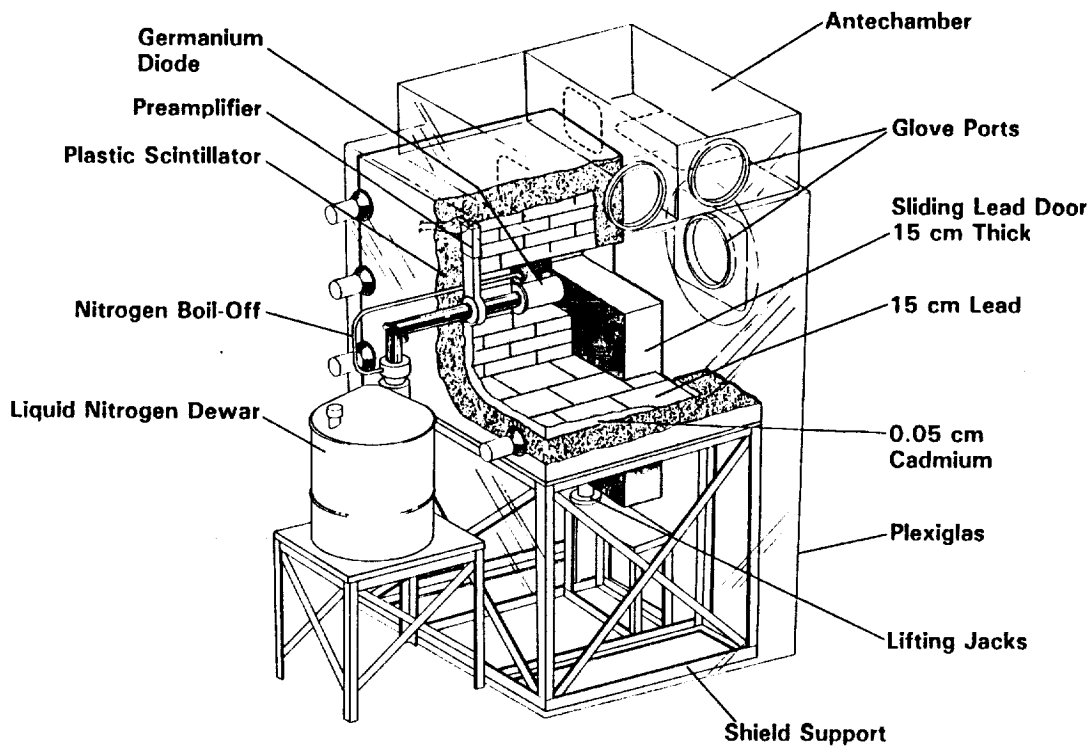


Figure 1. Antic cosmic-shielded, above-ground, ultra-low-background 31.5% HPGe gamma-ray spectrometer.

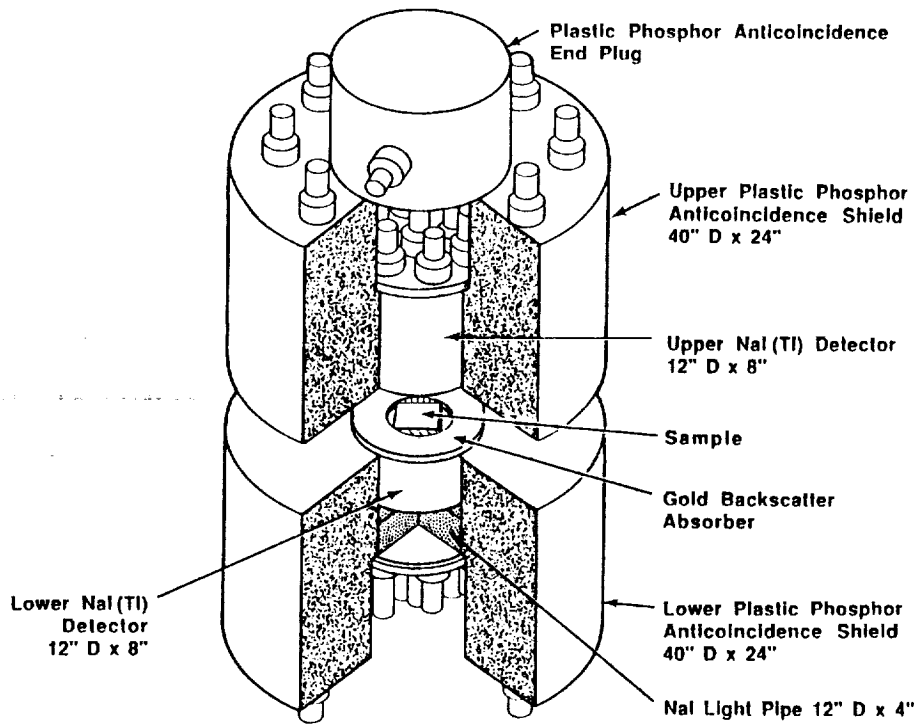


Figure 2. A large detector, anticoincidence shielded multidimensional NaI(Tl) gamma-ray spectrometer.

Table 1. Comparison of Counting Efficiencies and System Backgrounds for NaI(Tl), 31.5% HPGe and 90% HPGe Spectrometers for  $^{60}\text{Co}$  in 5.08-cm Square by 0.32-cm Thick Cobalt Metal Samples.

|   | <u>NaI(Tl) System</u> | <u>31.5% HPGe System</u> | <u>90% HPGe System</u> |
|---|-----------------------|--------------------------|------------------------|
| Counting Efficiency<br>(counts per gamma-ray) | 0.133                 | 0.030                    | 0.097                  |
| System Background<br>(counts per day)         | 34                    | 6                        | 14                     |

Table 2. Activity of  $^{60}\text{Co}$  in 5-cm Square Steel and Cobalt Samples.

| <u>Identification</u> | <u>NaI(Tl) System<br/>(pCi/kg)</u> | <u>31.5% HPGe System<br/>(pCi/kg)</u> |
|-----------------------|------------------------------------|---------------------------------------|
| Cobalt                |                                    |                                       |
| I-C9                  | $33.9 \pm 2.1$                     | $29.4 \pm 2.9$                        |
| G12-A-1-F-N           | $34.1 \pm 2.1$                     | $27.4 \pm 2.7$                        |
| I-H12 (A+B)           | $208 \pm 11.3$                     | $204 \pm 20$                          |
| Stainless Steel**     |                                    |                                       |
| LHDS2                 | $0.84 \pm 0.30$                    | $0.9 \pm 0.4$                         |
| LHDS3                 | $1.3 \pm 0.2$                      | N/A                                   |
| LHDS5                 | $1.2 \pm 0.2$                      | N/A                                   |
| LHDN2                 | $1.3 \pm 0.3$                      | N/A                                   |
| LHDN3                 | $1.3 \pm 0.2$                      | N/A                                   |

\*\* For the stainless steel samples, the sample blank was derived from stock other than that used in the actual LDEF samples; low inherent  $^{60}\text{Co}$  activity in the blank would result in an artificially high activity.

by  $^{60}\text{Co}$ . For purposes of comparison, the same source positions are used relative to the center of the face of the end cap of the germanium detector and the center of the face of one NaI(Tl) detector. All measurements are normalized to 1.00 at this position for each detector system. The relative efficiencies are summarized in Table 3. In general, the absolute efficiency of the NaI(Tl) system is greater than that of the HPGe system.

Measurements for one set of data in Table 3 were taken with no absorbing material between the source and the detector and show the relative efficiency decrease as the source is moved away from the center of the HPGe detector end cap. The relative efficiency for the NaI(Tl) spectrometer also decreases as the source is moved away from the axial center of the crystals. The greatest efficiency for the HPGe spectrometer occurs when the source is centered on the face of the detector end cap, since the greatest solid angle is subtended at such a position. The relative counting efficiency with the HPGe detector is reduced by a factor of 2.7 with the source 1.27 cm away from the end cap and offset 5.08 cm laterally whereas the data in Table 3 indicate only minor variations of between 4 and 6% with the NaI(Tl).

Measurements for the second set of data in Table 3 were taken with an aluminum sample and show that the relative efficiency decreases more severely due solely to attenuation of the gamma rays by the aluminum as the source is moved away from the HPGe, up to a factor of 4 when the source is located 1.27 cm away and offset 5.08 cm; however, with the same offsets the relative efficiency of the NaI(Tl) spectrometer actually increases by 16% with the  $^{22}\text{Na}$  source and by 2% with the  $^{60}\text{Co}$  source. This increase in relative efficiency is due to an edge effect. When the source is located near the edge of the aluminum, more gamma rays can escape the source and be absorbed in the two NaI(Tl) crystals because they pass through only a very thin section of aluminum as shown in Figure 3.

## EXPERIMENTAL RESULTS

The LDEF aluminum samples analyzed at PNL for  $^{22}\text{Na}$  consisted of rectangular slabs of aluminum of various dimensions. The slabs were centered on the face of the lower NaI(Tl) crystal for counting (this geometry having been previously standardized for samples of similar sizes and composition). The counting times were typically several days. Uncertainties reported include counting statistical uncertainties plus 5% for the combination of absolute standardization uncertainty, geometrical parameters such as the edge effect, and weighing errors. The results are shown in Table 4.

Sample KP-6 was also counted on the HPGe system. The count rate for the 511-keV peak plus the 1274.5-keV peak was about half the count rate observed with the NaI(Tl) system. If only the 1274.5-keV peak were used, then the count rate would be about 5% that seen with the NaI(Tl) system.

## DISCUSSION

Results from both detector systems, as shown in Table 2, indicate that the activity in cobalt sample I-H12 (A+B) is 6 times higher than the activity measured in the other cobalt samples. The higher activity is likely due to higher thermal-neutron fluence. Such a likelihood is further discussed by Alan Harmon in the proceedings of this symposium (ref. 5).

Table 3. Comparison of the Relative Efficiencies of HPGe vs. NaI(Tl) Spectrometers for a 9.5-mm Diameter Source Positioned in Various Locations Relative to a Sample 1.27-cm Thick By 8.255 cm Square.

| Source Location (cm)  |               | Relative Efficiency |                      |                   |                      |
|-----------------------|---------------|---------------------|----------------------|-------------------|----------------------|
| Distance from Surface | Radial Offset | No Absorber         |                      | Aluminum          |                      |
|                       |               | 31.5 % HPGe Diode   | NaI(Tl) Spectrometer | 31.5 % HPGe Diode | NaI(Tl) Spectrometer |
| 0                     | 0             | 1.00                | 1.00                 | 1.00              | 1.00                 |
| 0                     | 2.54          | 0.80                | 0.99                 | 0.80              | 1.01                 |
| 0                     | 5.08          | 0.40                | 0.94                 | 0.40              | 1.13                 |
| 1.27                  | 0             | 0.70                | 1.02                 | 0.56              | 1.03                 |
| 1.27                  | 2.54          | 0.58                | 0.99                 | 0.46              | 1.04                 |
| 1.27                  | 5.08          | 0.38                | 0.96                 | 0.25              | 1.16                 |

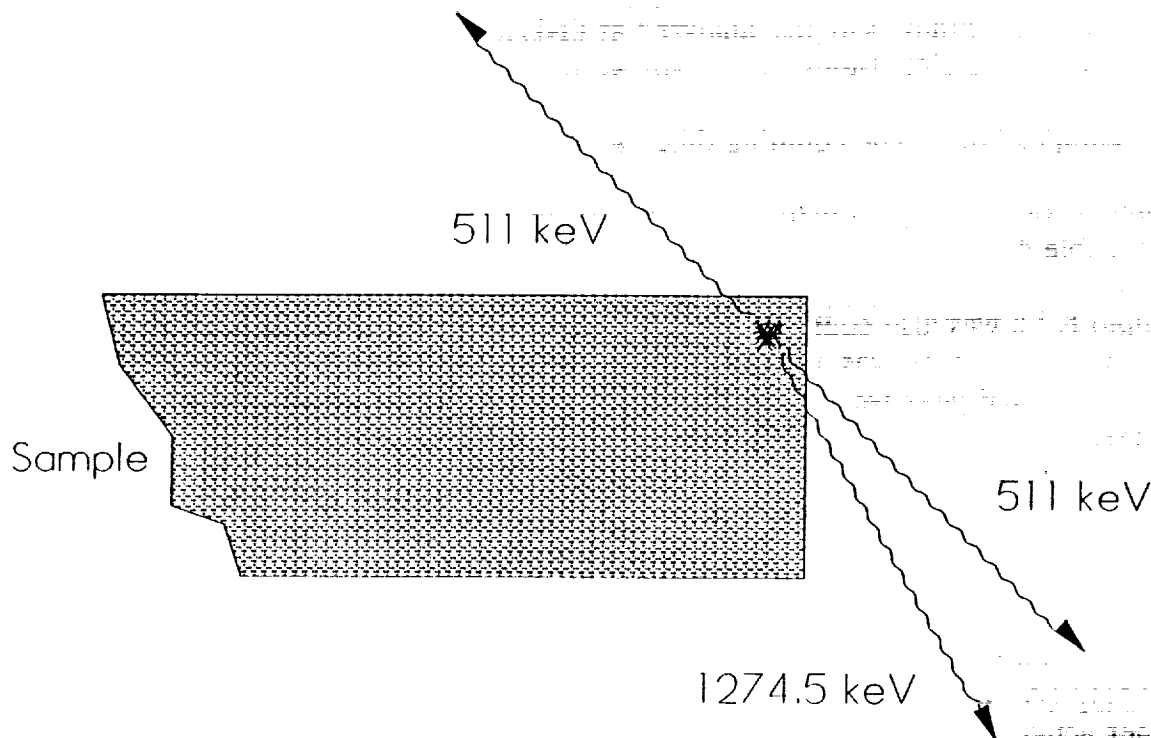


Figure 3. Schematic illustration of the edge effect for the decay of  $^{22}\text{Na}$ .

Table 4. Activity of  $^{22}\text{Na}$  in LDEF Aluminum Slabs as Determined Using a Large NaI(Tl) Detector Spectrometer.

| <u>Identification</u> | <u>Weight (kg)</u> | <u>Thickness (g/cm<sup>2</sup>)</u> | <u>Counting Efficiency (%)</u> | <u><math>^{22}\text{Na}</math> Activity</u> |   |
|-----------------------|--------------------|-------------------------------------|--------------------------------|---|---|
|                       |                    |                                     |                                | <u>(pCi/kg)</u>                             | <u>Uncertainty (1<math>\sigma</math>)</u> |
| KP-6                  | 0.06533            | 2.532                               | 18.81                          | 129.0                                       | 6.6                                       |
| KP-7                  | 0.06533            | 2.532                               | 18.81                          | 120.2                                       | 6.2                                       |
| CA-W-EAN-1            | 0.02203            | 0.535                               | 21.98                          | 106.3                                       | 5.7                                       |
| CA-W-EAN-2            | 0.04994            | 1.261                               | 21.10                          | 95.4  | 4.9                                       |
| CA-W-EAN-3            | 0.07078            | 1.938                               | 19.86                          | 82.6  | 4.4                                       |
| CA-W-EAN-4            | 0.07748            | 2.527                               | 18.81                          | 78.7  | 4.0                                       |
| CA-W-EAN-5            | 0.06715            | 2.376                               | 19.07                          | 70.3  | 3.7                                       |
| CA-W-SPS-1            | 0.02117            | 0.514                               | 22.03                          | 101.5                                       | 5.9                                       |
| CA-W-SPS-2            | 0.04949            | 1.254                               | 21.11                          | 89.8  | 4.6                                       |
| CA-W-SPS-3            | 0.07034            | 1.926                               | 19.88                          | 83.2  | 4.4                                       |
| CA-W-SPS-4            | 0.07590            | 2.476                               | 18.89                          | 74.7  | 3.9                                       |
| CA-W-SPS-5            | 0.06532            | 2.501                               | 18.85                          | 68.8  | 3.7                                       |

HPGe detectors can be readily calibrated using homogeneous standards, even for radionuclides with coincident emissions, such as  $^{22}\text{Na}$  which emits a 0.55-MeV  $\beta^+$ , but errors introduced from coincidence summing can be significant and some method of correction must be used (ref. 1). However, there is no reliable method for calibrating HPGe detectors for situations where the activity is not uniformly distributed throughout the sample. Furthermore, the range of the  $\beta^+$  in the sample material will affect the location at which annihilation occurs, especially in thin samples. Since coincidence summing is actually used for the measurements with the NaI(Tl) system, and due to the low sensitivity to source position, errors are vastly reduced even for inhomogeneous samples.

Measurements on samples of varying shapes and sizes requiring high precision are sometimes required and uncertainties due to edge effect must be minimized. For homogeneous samples, efficiency calibration with standards of similar shape and surface density are sufficient. However, for highly inhomogeneous samples, a better method must be used. One such method is to include a small border of blank material around the perimeter of the sample. The correct thickness and extent of the border material can be determined from mathematical calculations of absorption parameters. Uncertainties from edge effect have little significance for large samples

counted on small HPGe detectors since the edge of the sample extends past the outer surface of the HPGe crystal. However, for the large HPGe detectors currently available, the edge effect may contribute a significant uncertainty for 1-cm thick by 5-cm square stainless steel samples or other relatively thick, high density samples.

## SUMMARY

Large NaI(Tl) detector systems as described above are competitive with, and in many cases superlative to, even the largest low-background HPGe detector systems for measuring radionuclides which decay with coincident gamma rays of at least moderate energies. The combination of efficiency and system background are at least comparable to the best HPGe systems operating above ground. Large NaI(Tl) detector systems are especially advantageous for large inhomogeneous samples because of their low sensitivity to variable source position.

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