Si(Li)-NaI(Tl) SANDWICH DETECTOR ARRAY FOR MEASUREMENTS
OF TRACE RADIONUCLIDES IN SOIL SAMPLES

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An ultra-sensitive x-/γ-ray detector system for assaying trace radioactivity in actinide contaminated soil and ash samples has been developed. The new system consists of an array of 6 large Si(Li) x-ray detectors sensitive on both faces and mounted on edge in a paddle-shaped cryostat with a 14 cm dia. Be window on each side. The paddle, with a sample of the soil placed at each window, is sandwiched between 2 large NaI(Tl) scintillators which suppress the γ background. With x rays being measured simultaneously from soil in 2 sample holders and background reduced by 50% using anticoincidence, the sensitivity of this detector is 4 times higher than that of conventionally mounted Si(Li) detectors. A soil sample containing 50 pCi/g ²³⁹Pu was measured in 5 min with an uncertainty of <20% and a sample containing 7 pCi/g was measured in 1 hr. With FWHM resolution of 400 eV at 17 keV, the U_Lβ₁ and Np_Lβ₁ x-ray peaks are resolved thus permitting measurement of trace Pu in the presence of Am-241. This is the most sensitive and selective detector known for nondestructive assay of radioactivity in soil and other samples.

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I. INTRODUCTION

Radionuclide assay of soil, ash or sediment samples by photon spectrometry is far less laborious and hence, less costly than radiochemical analysis. Moreover, with x-ray spectrometry assay of several actinides and fission fragments can be made in a single measurement, whereas radiochemical analysis usually requires several sequential procedures for each preselected radionuclide, whether it is present in the sample or not. However, radioactive contaminants of great interest, such as plutonium, often have only very weak γ -ray emission and therefore γ rays are poorly suited for measurements of low-level activity. Sherman, et al¹ have shown that for several Pu isotopes the expected count rate of $UL\beta_1$ x rays emanating from a soil sample is 10-100 times higher than that of the γ rays and hence these x rays are better suited for low-level measurements. Our objective in this work was to develop the capability to detect incremental increases in soil activity, due to contaminants, comparable to the natural activity. This would require a detection sensitivity in the range of 1 pCi/g. This paper describes the design and performance of a Si(Li)-NaI(Tl) detector system in which this sensitivity has been achieved. This system is the culmination of extensive R&D in large area Si(Li) detector systems. The development of this spectrometer involved a comprehensive study of actinide x-ray spectroscopy and soil assay methods including the composition and level of natural radioactivity in soil. Other systems, principally using Ge rather than Si detectors, have been developed ²⁻⁵ for low-energy photon spectrometry; however, for soil assay, the Si(Li)-NaI(Tl) spectrometer offers a significant increase in sensitivity.

Total α activity cannot be determined by x-ray spectrometry without prior knowledge of the isotopic composition. This is usually determined by mass spectrometry, or as more recently reported, by measurement of Lx/ α -ray activity ratio.⁶ These methods require chemical processing of the soil sample and are therefore not as convenient for soil analysis as direct photon spectrometry.

Strauss, et al⁷ have shown that the isotopic ratio in soil samples can be determined, in the case of ^{239,240} Pu, for example, when measurement of the low energy γ rays emitted in the parent decay is made in a few selected samples. Although the emission of these γ rays is weak and the uncertainty may be large, due to the "Peter-Paul Effect", the uncertainty in the total activity is small.

II. DETECTOR DESIGN

Detector sensitivity is inversely proportional to the time t required to attain a given statistical precision. It can be shown that

$$t \propto \frac{1}{p} \left(1 + \frac{2}{p/b} \right) \quad (1)$$

where p and b are the peak and background count rates respectively. High sensitivity implies a capability to measure low activity in a short time. In order to achieve this capability, the detector should be designed to maximize p and p/b . The peak count rate p can be maximized by 1) using a detector having a large sensitive area and high detection efficiency for the photons of interest and 2) by using good detector-to-sample geometry. The peak-to-background ratio p/b can be maximized by 1) using a detector having high energy resolution and low sensitivity to background radiation and 2) by suppressing whatever background is sensed. All of the above criteria have been used in the design of the present detector system.

The principal detector consists of a circular array of 6 large Si(Li) x-ray detectors (34 mm dia x 7 mm thick). The detectors have thin windows on both faces⁸ and are mounted on edge in order to count two samples of the soil simultaneously. They are mounted in a paddle-shaped cryostat with a Be window on each side as shown schematically in Fig. 1. The paddle, with a sample of the soil placed at each Be window, is sandwiched between two large NaI(Tl) scintillators* operating in anticoincidence with the Si(Li) detectors to

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suppress the γ background. The Si(Li) detector array is shown separately in Fig. 2 and sandwiched between the NaI(Tl) scintillators inside a radiation shield in Fig. 3.

Silicon detectors are preferable to Ge detectors because Si(Li) detectors combine high energy resolution with low sensitivity to γ background as discussed by Sherman, et al.⁹ The negligible K x-ray escape probability in Si is also an important advantage of Si vis-a-vis Ge.

The objective in the design of this system was to maximize detection efficiency for given size entrance windows and anticoincidence scintillators. Hence, considerable technology was brought to bear to restrict the thickness of the paddle-shaped cryostat (25 mm). To maximize the detector aperture, a single large Be window, instead of small individual windows for each crystal, was used on each side. The 14 cm dia., 0.5 mm thick window extends 1 cm beyond the array perimeter thus permitting the use of large-area samples. One of the windows is demountable to allow easy access to the internal electronic components and Si(Li) crystals. Each detector was passivated¹⁰ with amorphous Si to produce a stable surface which permitted repeated temperature cycling and thus facilitated the assembly of the array. A detailed description of the detector system design is in preparation¹¹.

The sample holder is made up of a 5 mm thick lexan ring having an inside diameter of 14.5 cm. A 0.12 mm thick mylar window is cemented to each side of the ring. The space between the two mylar sheets is filled with ~100 g of soil and sealed at slightly negative pressure to prevent the soil from shifting. The sample thickness of 5 mm corresponds to approximately two mean free paths for the ULy x rays (20 keV). It has been shown¹² that this thickness yields essentially the full x-ray intensity available from the sample.

The signal processing system consists of a cooled-FET preamplifier with transistor reset¹³ followed by a pulse-shaping amplifier¹⁴ used with a peaking

time of 70 μ sec. The outputs from the six amplifiers are multiplexed[†] into a single ADC and routed to separate memory regions in the computer.

III. DETECTOR PERFORMANCE

Energy resolution is a key parameter for detector sensitivity. It determines the peak-to-background ratio p/b as shown in eq. 1 and also the extent to which x-ray peaks due to contaminants can be separated from those due to the natural activity in soil. In addition to sensitivity, the resolution affects the capability to assay several radionuclides simultaneously. Thus, for example, Pu is often found in combination with ^{241}Am and in some instances also with ^{237}Np . The associated x rays are $\text{UL}\beta_1$ (17.22 keV), $\text{NpL}\beta_1$ (17.75 keV), and $\text{PaL}\beta_1$ (16.69 keV). The resolution of these peaks was a principal consideration in the design of this detector system. This requires a FWHM resolution less than 500 eV at 17 keV.

The $\text{L}\beta_{1,3,5}$ is a peak complex and therefore the peak width is not a very sensitive measure of detector resolution. In the case of ^{241}Am , for example, the $\text{NpL}\beta_3$ (17.99 keV) component is 20% of the $\text{NpL}\beta_{1,3,5}$ peak. We therefore used the peak-to-valley P/V ratio as a measure of detector resolution, where P is the point of highest intensity in the $\text{NpL}\beta_{1,3,5}$ peak and V is the point of lowest intensity in the valley between that peak and the $\text{NpL}\beta_2$ (17.25 keV) peak. For the detectors in this array the P/V ratios are between 7.5 and 5, corresponding approximately to a pulse generator FWHM resolution of 300-400 eV respectively. The FWHM of the 26.4 keV γ ray from ^{241}Am measured with the different detectors in the array is in the range of 400-500 eV.

The benefit of high detector resolution is seen in the x-ray spectrum of a contaminated soil sample containing 77 pCi/g $^{239,240}\text{Pu}$ and 7.1 pCi/g ^{241}Am (Fig. 4). The $\text{UL}\beta_1$, $\text{NpL}\beta_1$ and the $\text{UL}\gamma_1$, $\text{NpL}\gamma_1$ peaks are resolved thus permitting direct assay of Pu and Am.

[†] EG&G ORTEC, Oak Ridge, Tennessee (Model 476)

One measure of detector sensitivity is the counting time required for a given statistical precision (eq. 1). An x-ray spectrum of soil containing 50 pCi ^{239}Pu and 6 pCi ^{241}Am is shown in Fig. 5. A counting time of 5 min with the 6-detector array gave a relative $\sigma = 18\%$ for the Pu. Since the data from the 6 detectors are recorded separately and are not readily combined, we show, for the sake of illustration, in Fig. 5 an equivalent spectrum from a single detector obtained in 30 min.

Another measure of sensitivity for soil assay is the minimum contaminant activity detectable in the presence of the natural activity. An x-ray spectrum of soil containing 6.8 pCi ^{239}Pu is shown in Fig. 6. A 60 min measurement with the array gave a relative statistical uncertainty $\sigma = 17\%$. The spectrum shown in the illustration in Fig. 6, obtained with a single detector in 360 min, is equivalent to that obtained with the array in the 60 min measurement. The RaL and $\text{ThL}\beta_1$ peaks are produced in the natural U and Th decay chains. Note that the intensity of the $\text{ThL}\beta_1$ peak is higher than that of the $\text{UL}\beta_1$ peak due to the Pu, however the $\text{UL}\beta_1$ peak is clearly above the continuum. Thus, the minimum detectable activity of Pu in soil is less than 6.8 pCi/g.

The capability of this type detector system to distinguish trace radioactivity from a high background is illustrated in the spectrum of an ashed fecal sample containing 15 pCi $^{239,240}\text{Pu}$ (Fig. 7). Since the sample size was limited to 7.4 g, only one of the Si(Li) detectors in the array was used in combination with the NaI(Tl) anticoincidence detectors. Anticoincidence reduced the continuum by $\sim 50\%$. Without this reduction the $\text{UL}\beta_1$ peak in Fig. 7 would barely be discernible. The sensitivity exhibited in this measurement corresponds to an activity level of 2 pCi/g $^{239,240}\text{Pu}$.

Background suppression reduces the counting time required for a given statistical precision. The extent of this reduction with the present system was determined from eq. 1 and from measurements of several soil samples when anticoincidence was used and not used (Fig. 8). It is seen that for a 2 pCi/g

^{239}Pu measurement, anticoincidence background suppression reduces the counting time by 50% for $\sigma = 20\%$. For a 20 pCi/g sample the reduction is 30%. Thus, background suppression is seen to be most valuable for low level measurements.

IV. DISCUSSION

The background reduction in soil¹ obtained with these NaI(Tl) anticoincidence scintillators is ~50% in the 15-25 keV range. A significant fraction of the remaining continuum is believed to be due to photons degraded by Compton scattering in the soil matrix and subsequently fully absorbed in the Si(Li) detectors. These degraded photons are not amenable to rejection by anticoincidence; thus a closer geometry, which would increase the solid angle subtended by the NaI(Tl) scintillators, is not likely to reduce the background substantially.

This detector system can be used for measurements of samples other than soil as was shown in Fig. 7 for the case of a fecal sample, and in 1980 for the ash from Mount St. Helens following its eruption.¹⁵ When measurements are made of samples consisting of different matrix materials, one must consider the x-ray attenuation through the matrix and the sample geometry. The number of x rays emanating from the sample is inversely proportional¹² to the mass attenuation coefficient μ/ρ . Thus, when comparing x-ray intensities of a given sample to that of soil, it is necessary to measure the mass attenuation coefficients¹⁵ of the samples and correct for the relative absorption losses.

V. CONCLUSION

There is considerable merit in mounting a Si(Li) detector on edge. It doubles the sensitive area of the detector and it permits anticoincidence background suppression. With x rays being measured simultaneously from 2 samples of the soil and background reduced by 50%, the sensitivity of an edge-mounted detector is 4 times higher than that of a conventionally mounted Si(Li) detector. In earlier studies^{1,7,15} the capabilities for measuring radioactivity in soil have been explored by using a single edge-mounted detector. In the

present study we demonstrate the increased sensitivity obtained with an array of 6 edge-mounted Si(Li) detectors in close geometry with a single large Be window over each face of the array. The use of background suppression by anticoincidence increases the sensitivity further. For an activity level of 1-2 pCi/g ^{239}Pu , anticoincidence reduces the measurement time by 50%. It is shown that incremental increases of trace activity, due to contaminants, comparable to the natural activity in soil can be measured with this detector array to 20% precision in an overnight run.

A modified version of this Si(Li)-NaI(Tl) detector array is now being developed for measurements in vivo of internal body actinide depositions.

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Captions

- Fig. 1 Configuration of the Si(Li)-NaI(Tl) x-/γ-ray spectrometer. The detector system consists of a circular array of 6 edge-mounted Si(Li) x-ray detectors each having two sensitive faces. The array, with a sample of the soil placed at each Be window, is sandwiched between 2 large NaI(Tl) anticoincidence scintillators which suppress the γ background due to the radioactivity in the soil.
- Fig. 2 Photograph of the paddle-shaped cryostat housing the Si(Li)-detector array shown schematically in Fig. 1. The paddle is 25 mm thick and has a 14 cm dia., 0.5 mm thick Be window on each face.
- Fig. 3 Photograph of the Si(Li)-NaI(Tl) detector system inside a radiation shield as shown schematically in Fig. 1. The Si(Li) detector array in the paddle-shaped cryostat (Fig. 2), with a sample of the soil on each side, is sandwiched between two large NaI(Tl) scintillators each of which is viewed by an array of 6 photomultipliers.
- Fig. 4 X-ray spectrum from a contaminated soil sample showing the energy resolution of the Si(Li) detector system. The UL and NpL x rays are due to the decay of Pu and Am respectively. The two $L\beta_1$ x-ray peaks, 0.5 keV apart, are resolved thus providing the specificity required for separate assay of radionuclides having closely spaced x-ray peaks.
- Fig. 5 X-ray spectrum from a contaminated soil sample reflecting the sensitivity of the spectrometer in the assay of Pu in the presence of Am. The UL and NpL x rays are due to the decay of Pu and Am respectively. Counting 5 min with the 6-detector array gave a relative $\sigma = 18\%$ for a measurement of a 50 pCi/g Pu sample (UL β_1 peak contains 60 counts net). The above spectrum, obtained for this illustration with a single detector in 30 min, is equivalent to that obtained with the array in the 5 min measurement.

Fig. 6 X-ray spectrum from a contaminated soil sample reflecting the sensitivity of the spectrometer in detecting trace Pu in the presence of the natural activity. The UL x rays are due to the decay of Pu, the RaL and ThL x rays are due to the decay of natural U and Th. Counting 60 min with the 6-detector array gave a relative $\sigma = 17\%$ for a measurement of a 6.8 pCi/g Pu sample (UL₉₁ peak contains 80 counts net). The above spectrum, obtained for this illustration with a single detector in 360 min, is equivalent to that obtained with the array in the 60 min measurement. Note that the minimum detectable activity of Pu in soil is less than 6.8 pCi/g.

Fig. 7 X-ray spectrum from a fecal sample containing 15 pCi Pu. Only one of the Si(Li) detectors in the array was used. Anticoincidence reduced the continuum by ~50% thus allowing a detectable Pu activity level of 2 pCi/g.

Fig. 8 Counting time vs. Pu activity in soil for a relative statistical precision of $\sigma = 20\%$. When the Pu activity approaches the natural activity in soil (~1 pCi/g), anticoincidence reduces the required counting time substantially.

Fig. 1

EML S1839

77 pCi/g $^{239.240}\text{Pu}$

7.1 pCi/g ^{241}Am

Fig. 5

Fig. 6

Fig. 7

Fig. 8