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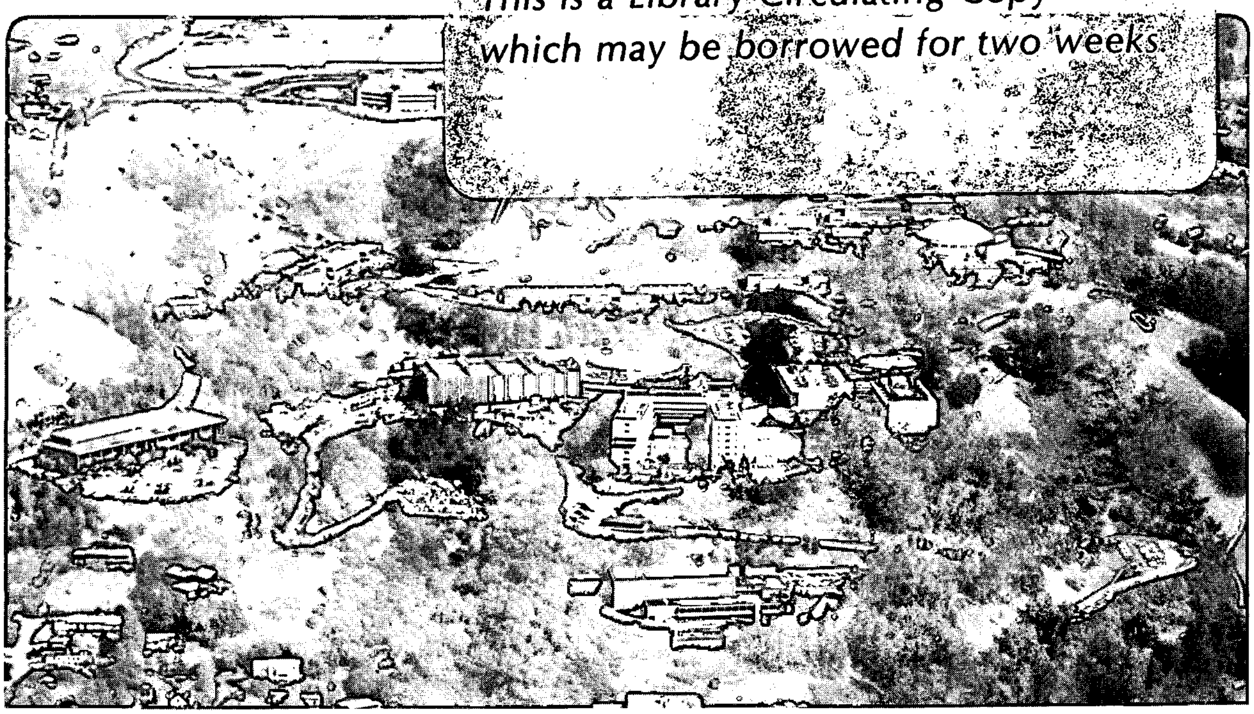

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ABSTRACT

A system containing six cooled, 34 mm diam by 7 mm thick, high-resolution Si(Li) detectors designed to maximize the sensitivity for counting X rays in the 10-30 keV range to measure trace radionuclides in soil samples has been successfully fabricated. The detectors were mounted in a paddle-shaped cryostat with a single large beryllium window on each side. This configuration provides for efficient anticoincidence background suppression and effectively doubles the sensitive detector area because X rays can impinge on the detectors from both sides. To maximize detection efficiency, the thickness of the cryostat was held to a bare minimum (25 mm); this caused severe difficulties during fabrication of the system. Cutting down the rim of the detectors reduced to an acceptable level the microphony caused by movement of the beryllium window that faces the lithium-diffused contact of the detectors. Since this system will be used for low level counting, careful testing was performed to select materials having the lowest radioactivity.

INTRODUCTION

The scientific goals associated with an ultra-sensitive X-/gamma-ray spectrometer system for assaying trace radioactivity in soil and ash samples contaminated with actinides have been chronicled in a series of previous papers.[1-6] An array of cooled large area Si(Li) detectors was designed to satisfy these goals. The present paper will discuss the technological developments made during the evolution of this system--a system that is the heart of the most sensitive and selective instrument available for nondestructive assay of radioactivity in soil and similar samples.

SILICON DETECTOR SYSTEM DESIGN

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

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

where t is the time required to attain a given statistical precision and 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 system should be designed to maximize p and p/b. The peak count rate can be maximized by: 1) using a detector having a large sensitive area and high detection efficiency for the photons of interest, and 2) using good detector-to-sample geometry. The p/b ratio can be maximized by: 1) using a detector having good energy resolution and low sensitivity to background radiation, 2) minimizing the presence of radioactivity in every component in the system, and 3) suppressing whatever background is sensed. These criteria have all been addressed in our design.

The principal detector system consists of a plane circular array of six large-area, high-resolution Si(Li) detectors mounted in a paddle-shaped cryostat with a beryllium window on each side, as shown schematically in Fig. 1. The cryostat, with a thin sample of the soil placed close to each beryllium window, is sandwiched between two large NaI(Tl) scintillators* operating in anticoincidence with the Si(Li) detectors to suppress the gamma background arising from Compton interactions. To maximize detection and gamma suppression efficiency for given size entrance windows and anticoincidence scintillators, the thickness of the paddle-shaped cryostat was held to a bare minimum (25 mm). This extremely restrictive configuration caused severe difficulties during fabrication of the system. A photograph of the cryostat is shown in Fig. 2 while Fig. 3 shows the cryostat sandwiched between the NaI(Tl) scintillators inside a radiation shield.

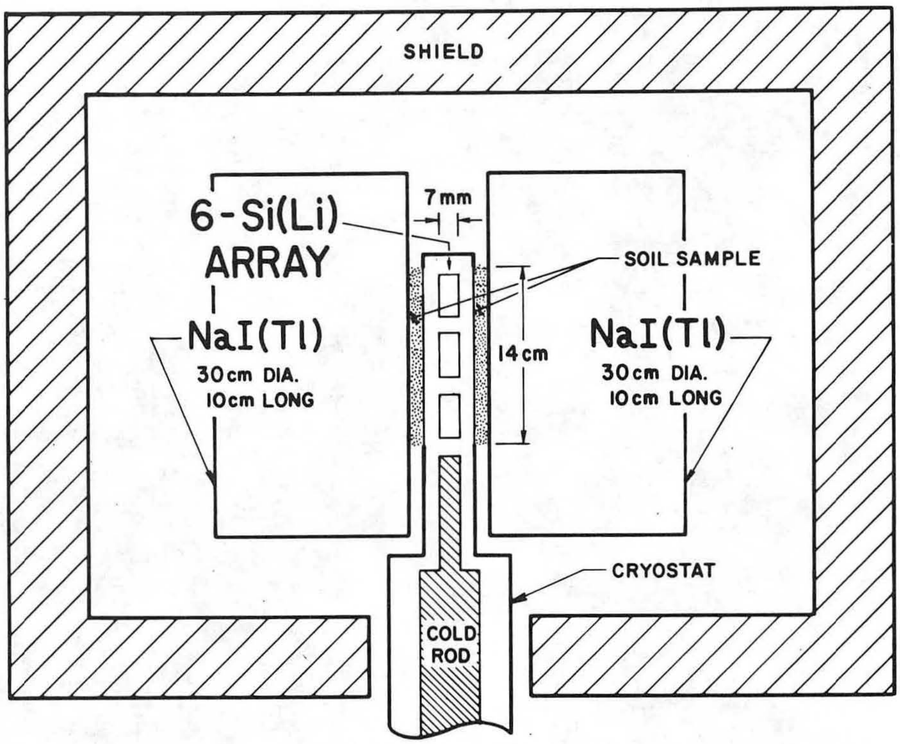

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Fig. 1 Configuration of the Si(Li)-NaI(Tl) X-/gamma-ray spectrometer. The principal detector system consists of a circular array of six large-area Si(Li) detectors mounted "on edge" so photons can impinge directly on the detectors from both sides. The array, with a sample of the soil placed at each beryllium window, is sandwiched between two large NaI(Tl) anticoincidence scintillators which suppress the gamma background arising from Compton interactions due to radioactivity in the soil.

* Harshaw/Filtrol, Solon, Ohio

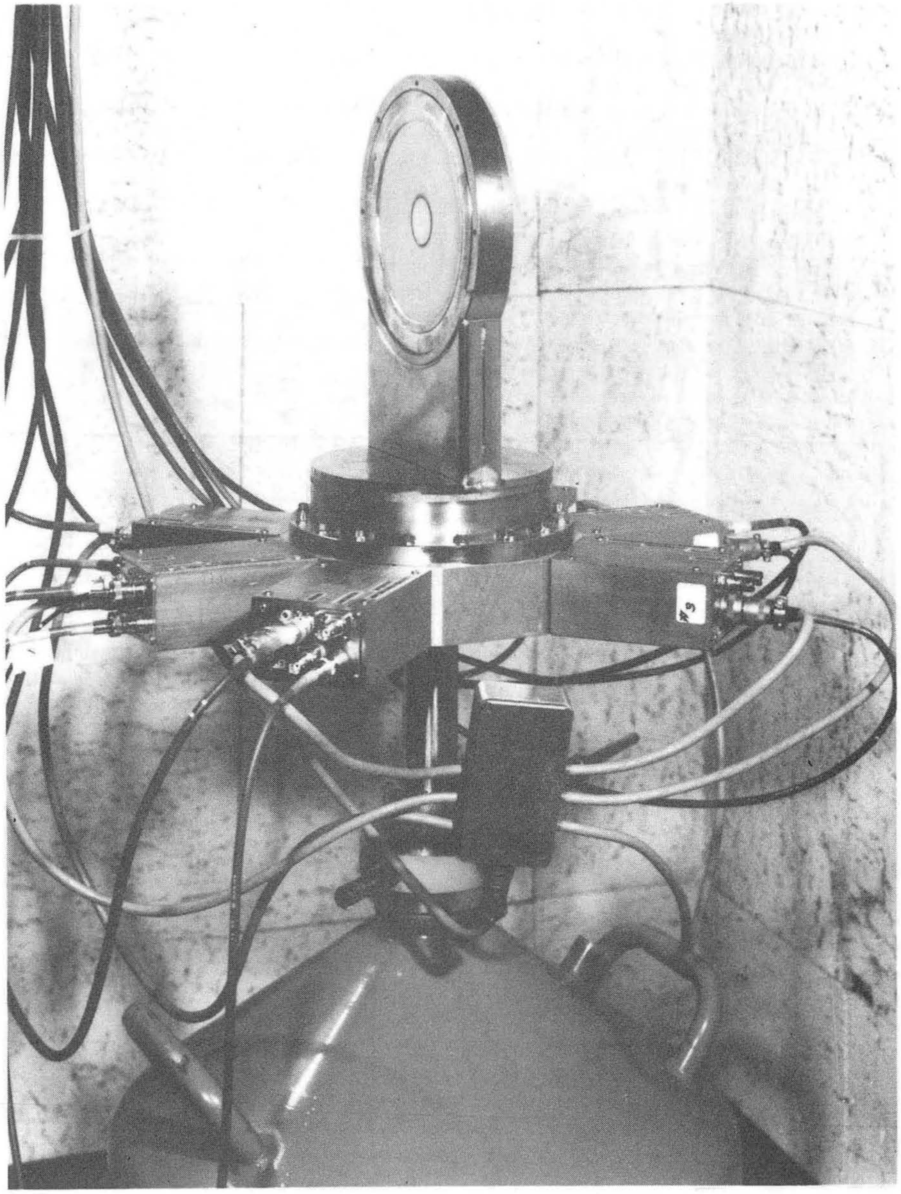

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Fig. 2 Photograph of the paddle-shaped cryostat housing the Si(Li)-detector array shown schematically in Fig. 1. The paddle is 25 mm thick and has a 14 cm diam, 0.5 mm thick beryllium window on each face.

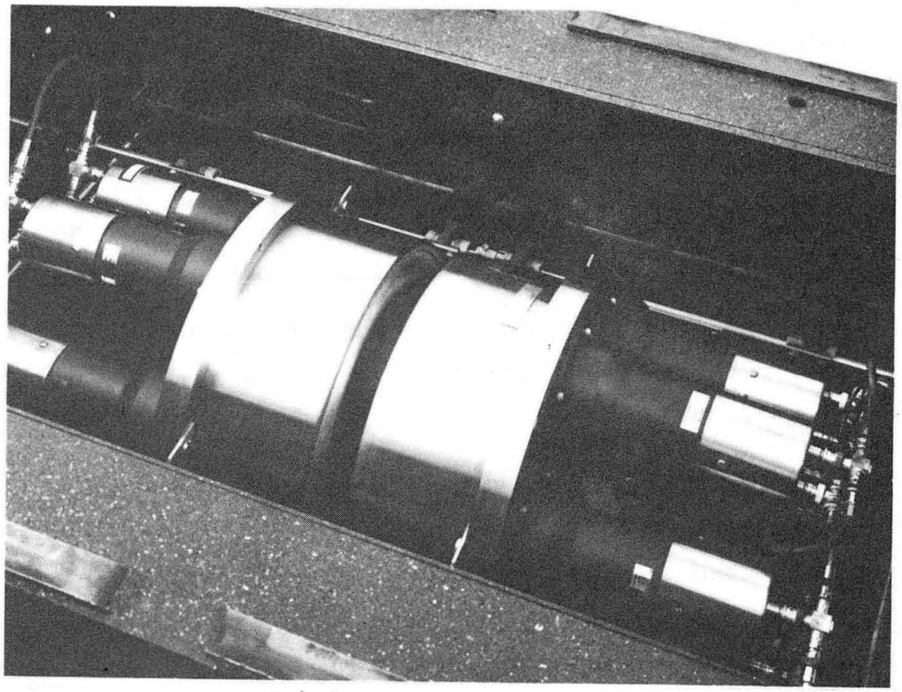

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Fig. 3 Photograph of the Si(Li)-NaI(Tl) detector system inside a radiation shield as shown schematically in Fig. 1. The Si(Li) detector array in the paddle-shaped cryostat (Fig. 2), with a sample of the soil on each side, is sandwiched between two large NaI(Tl) scintillators each of which is viewed by an array of six photomultipliers.

Energy resolution is a key parameter in detector sensitivity. It determines the peak-to-background ratio p/b as shown in Eq. 1 and is very important in determining the extent to which X-ray peaks due to contaminants can be separated from those due to the natural radioactivity 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 $UL_{\beta 1}$ (17.22 keV), $NpL_{\beta 1}$ (17.75 keV), and $PaL_{\beta 1}$ (16.69 keV). Separation of these peaks, which requires a FWHM resolution better than 500 eV at 17 keV, was a major consideration in the design of this detector system and particularly in determining the detector size.

To maximize the sensitivity for counting X rays in the 10-30 keV range, the design dimensions of each Si(Li) detector were chosen to be 34 mm diam x 7 mm thick. This choice was based on several considerations. To maximize the counting rate, detectors of the largest possible area would be preferable, and silicon crystals that would allow fabrication of detectors up to 70 mm in diameter are available. However, for a detector of a given thickness, the capacitance increases linearly with area causing a decrease in the signal-to-noise ratio. In principle, the capacitance of the detector could be kept constant by increasing the thickness to compensate for an increased area since the capacitance varies inversely with detector thickness. In practice, however, the maximum practical distance lithium can be drifted in silicon is about 8 mm; furthermore, detector sensitivity to gamma background increases with thickness. Faced with these limitations, and with our energy resolution goal in mind, we chose detector dimensions of 34 mm diam x 7 mm thick.

In the configuration used, the capacitance of each detector is about 16 pF; the corresponding pulser FWHM resolution at a peaking time of 35 μs is typically 350 eV using selected Siliconix U 311 FETs remounted in low loss packages. The leakage current of each detector is about 10^{-13} A at the operating temperature of about 90°K. The FET operating temperature of about 140°K is maintained by the dissipation of power in the FET (drain voltage of 3.5 V and drain current of 10 mA) in conjunction with the appropriate thermal barrier between the FET and the cold finger. The use of heaters to optimize the FET temperature in detector arrays (a technique commonly used in smaller systems) is undesirable because of the significant increase in heat load. A photograph of the copper cold finger, the silicon detectors and front-end electronics is shown in Fig. 4.

The signal processing system consists of a transistor reset preamplifier[7] followed by a pulse-shaping amplifier[8] used with a peaking time of 35 or 70 μs. The outputs from the six amplifiers are multiplexed into a single ADC and routed to separate memory regions in the computer.

Since the $L_{\beta1,3,5}$ is a peak complex, the width of this peak is not a very sensitive measure of detector resolution. In the case of ^{241}Am the $NpL_{\beta3}$ (17.99 keV) component is 20% of the $NpL_{\beta1,3,5}$ peak. Therefore, as a measure of detector resolution, we use the peak-to-valley ratio, the peak being the point of highest intensity in the $NpL_{\beta1,3,5}$ peak and the valley the point of lowest intensity in the valley between that peak and the $NpL_{\beta2}$ (17.25 keV) peak. The peak-to-valley ratios for the detectors in this array are between 7.5 and 5. The FWHM resolution of the 26.4 keV gamma-ray peak from ^{241}Am is about 425 eV.

Mounting the detectors "on edge", i.e., so photons can impinge directly on the detectors from both sides, not only provides for anticoincidence background suppression but also effectively doubles the sensitive detector area. To

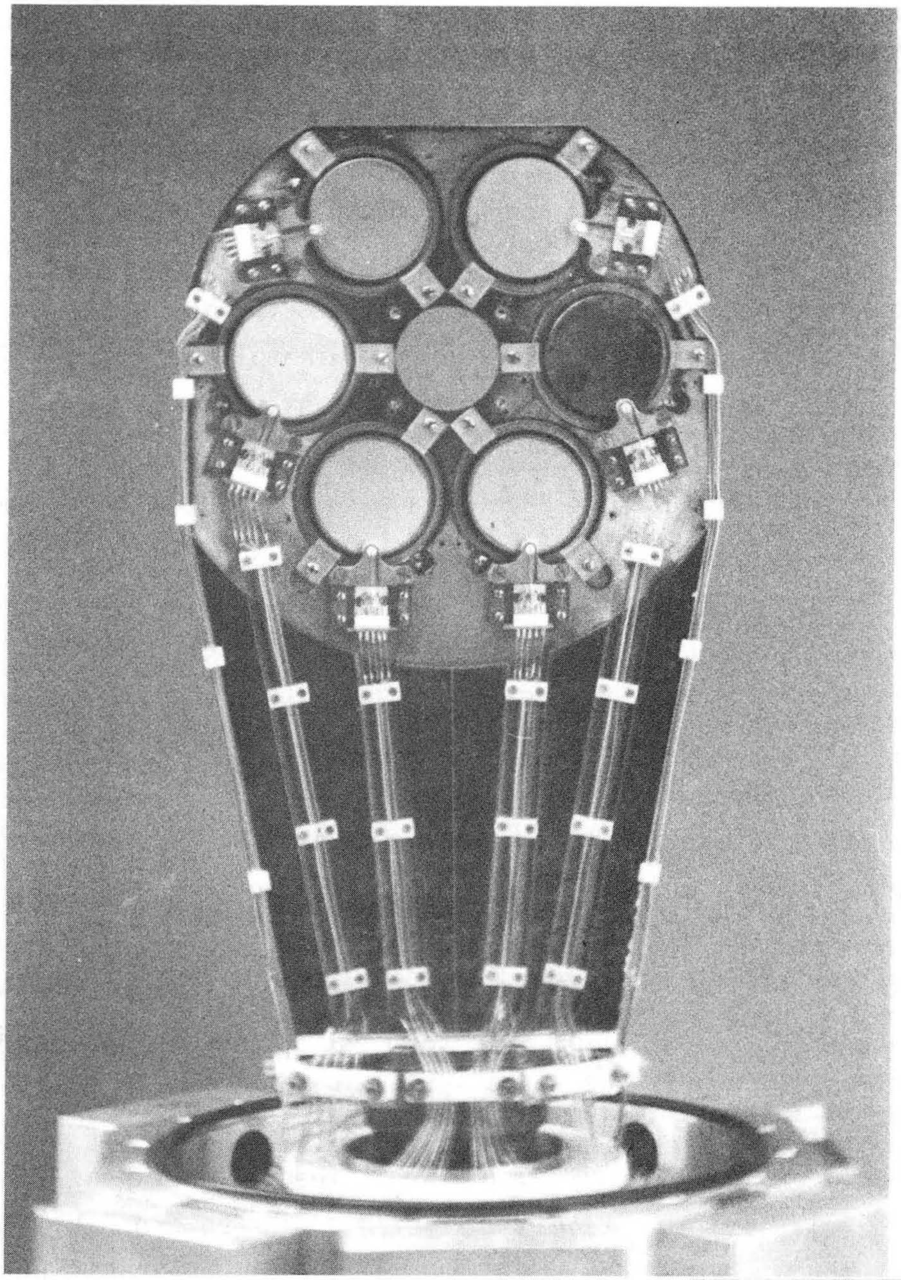

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Fig. 4 Photograph of the copper cold finger, the silicon detectors and front-end electronics.

achieve the same effective area with single-side entry would double the detector capacitance and the electronic noise would consequently increase by about 120 eV. Another significant benefit of the double-sided sensitivity is the diminished background because less cryostat material is required for the more compact system. Since photons impinge directly on the detectors from both sides, the lithium-diffused contact was made relatively thin, ~ 20 μm compared to the ~ 200 μm thickness typically found in thick lithium-drifted silicon detectors. Details of this thinning process have been reported previously.[9]

Removal of most of the rim of the detectors as shown in Fig. 5 was an important step in reducing the microphony in the system to an acceptable level. In a standard "grooved" Si(Li) detector (Fig. 5, left), the outer rim, which is at negative bias voltage (~ 1000 V), is close to the central lithium contact area that is connected to the very sensitive preamplifier input FET gate and, therefore, only a few tens of millivolts below ground potential. The FET gate is effectively not only the small wire but also the entire lithium contact area of the detector. This gate has capacitance to the beryllium window and to all other nearby materials including a capacitance to the rim of the detector, which we call the fringing capacitance. The fringing capacitance is of relatively little concern in most conventional spectrometers. However, in our system one beryllium window is very close to the lithium-diffused face of the detectors and extremely sensitive to sound-induced vibration. Movement of this beryllium window, which acts as a ground shield of a three-terminal capacitor, causes modulation of the fringing capacitance and the induced charge flow to the FET preamplifier results in very large microphonic signals. Cutting down the rim of each detector (Fig. 5, right) greatly reduces the fringing capacitance. This greatly reduces capacitance modulation due to window vibrations thereby substantially decreasing the microphony. The shallow rim also provides a convenient place to hold the detectors against the cold finger with clamps made of Lexan.

The benefit of good energy resolution is seen in the X-ray spectrum of a contaminated soil sample containing 77 pCi/g $^{239,240}Pu$ and 7.1 pCi/g ^{241}Am (Fig. 6). The $UL_{\beta 1}$, $NpL_{\beta 1}$ and the $UL_{\gamma 1}$, $NpL_{\gamma 1}$ peaks are sufficiently resolved to permit direct assay of Pu and Am.

As the number of detectors in a cryostat increases, the detector surface stability becomes increasingly important because of the additional handling and cycling each detector is subjected to. Hydrogenated amorphous silicon deposited in an RF plasma sputterer in an argon atmosphere containing 7% H_2 was used to passivate the surfaces of these detectors; these α-Si coatings produce very stable low-leakage surfaces without introducing additional noise when the detectors are operated close to liquid nitrogen temperature.[10]

The choice of silicon over germanium for this type of detector application has been discussed in detail in an earlier paper.[11] Briefly stated, silicon

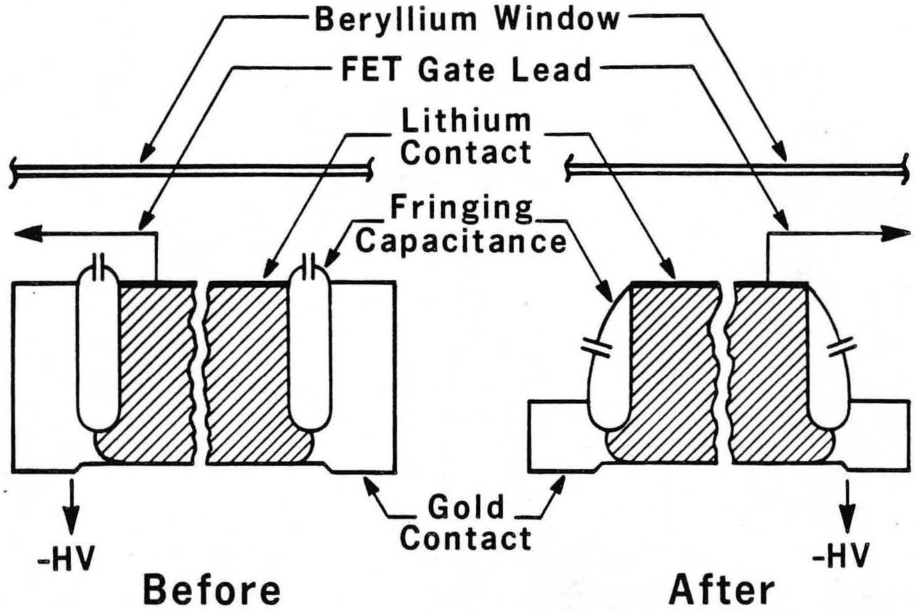

Fig. 5 The left portion shows a standard "grooved" Si(Li) detector with the beryllium window near the lithium-diffused contact. Movement of the window modulates the fringing capacitance which has ~ 1000 V across it. Cutting down the "high voltage" rim as shown on the right greatly reduces the fringing capacitance thereby substantially reducing the microphony. This modification also provides a convenient place to clamp the detector to the cold finger.

detectors are preferable because of their much lower sensitivity to gamma background and because the K X-ray escape probability is negligible in silicon detectors. An additional practical reason for the choice of silicon over germanium is that high-resolution germanium detectors must be shielded from infrared radiation whereas silicon detectors require no shielding. If germanium detectors were used, the thickness of the cryostat would have to be increased significantly to allow for a cold shield; this would conflict with a central design goal of minimizing the thickness of the cryostat.

BERYLLIUM ENTRANCE WINDOWS

To maximize the effective detector system aperture, a single large beryllium window, instead of small individual windows for each detector, was used on each side of the circular array. The 14 cm diam, 0.5 mm thick window extends 1 cm beyond the array perimeter thereby permitting the use of large-area samples. One of the windows is demountable to allow relatively easy access to the

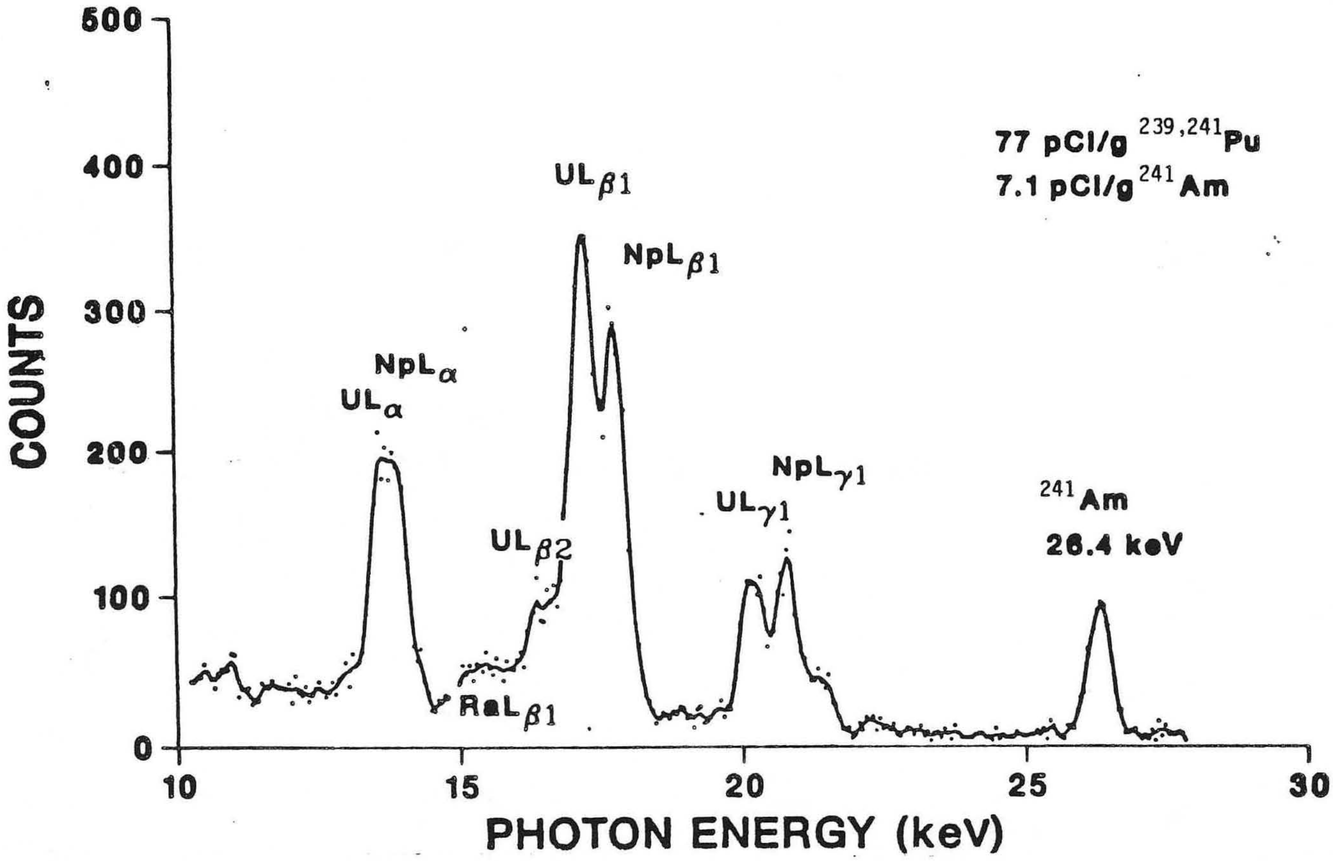

Fig. 6 X-ray spectrum from a contaminated soil sample. The UL and NpL X rays are due to the decay of Pu and Am, respectively. The two $L_{\beta1}$ X-ray peaks, 0.5 keV apart, are sufficiently resolved to permit direct assay of Pu and Am.

internal electronic components and detectors. To decrease the radiative heat load, aluminum was evaporated onto the inner (vacuum) surface of the beryllium to lower the emissivity.

Although a single large beryllium window provides significantly more transmission for low-energy photons from a diffuse source than separate windows over each detector, large beryllium windows present several serious problems. One of the major problems is finding beryllium sufficiently free of radioactivity. Since the uranium content in most of the available beryllium is far higher than desired for use in this system where a substantial quantity of beryllium is present near the detectors, a large number of samples had to be analyzed to find acceptable window material. Both X-ray fluorescence analysis and the LBL low-background gamma-ray counting facility were used to analyze the beryllium samples. The uranium content in the beryllium samples analyzed varied from < 2 ppm (our limit of measurement for these sheet-shaped samples) to 150 ppm; the uranium content in the beryllium used for the windows is < 2 ppm.

Attaching the beryllium window to the cryostat vacuum jacket was another major problem. The large area, thin window bows under vacuum causing great strain on the periphery of the window. This bowing also caused concern that the relatively brittle beryllium would eventually crack. In fact, during testing of one of our window designs a small crack developed in the center region of the window. This window was subsequently annealed, patched and used successfully in the system.

The cost of these beryllium windows is also very high (~ $4,000/window). The mechanical problems caused by the bowing could be reduced by using thicker beryllium windows, but the cost increases almost linearly with thickness. Furthermore, the additional material would increase the amount of uranium near the detectors.

We regard the use of these beryllium windows as only an interim solution to the problem of finding large-area windows that provide high transmission of relatively low-energy photons. This is especially true in situations where the presence of natural radioactivity is a concern. Consequently, we are currently experimenting with alternatives such as composites reinforced with Kevlar aramid.

CRYOSTAT MATERIALS

Since this system was to be used for low level counting, careful testing in the LBL low-background gamma-ray counting facility was performed on all materials being considered for use in the system to select materials having the lowest possible radioactivity. The copper cold finger, the stainless steel vacuum jacket surrounding the detector array and the beryllium windows comprise most of the material near the detectors. Oxygen-free high conductivity copper is the only structural metal consistently found to be free of activity to our limits of measurement and was the only material considered for the cold finger. We first fabricated the vacuum jacket from aluminum, but ultimately decided to switch to stainless steel because the slight ^{60}Co contamination in stainless steel was considered less of a problem in this application than the thorium contamination found in most aluminum. Furthermore, the additional strength of stainless steel significantly reduces the bowing of the vacuum jacket under vacuum.

CONCLUSION

A spectrometer consisting of a circular array of six large-area, high-resolution Si(Li) detectors mounted so photons can impinge directly on the detectors from both sides has been successfully fabricated despite great difficulties caused by the desire to restrict the thickness of the cryostat to an absolute minimum. The sensitivity of this system permits measurement of an activity level of 1 pCi of ^{239}Pu in 1 g of soil. The technology for fabricating these Si(Li) detector arrays may now be considered reasonably well developed. The major problem remaining is that of finding a better material for the large-area windows.

A modified version of this Si(Li) detector array is now being developed for measurements *in vivo* of internal body actinide depositions. In this system the space constraints are less severe and the X rays enter the detector from only one side.

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