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A COMPARISON OF LOW-ENERGY PHOTON DETECTORS FOR PLUTONIUM AND AMERICIUM WOUND COUNTING

THE DOW CHEMICAL COMPANY
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A COMPARISON OF LOW-ENERGY PHOTON DETECTORS FOR PLUTONIUM AND AMERICIUM WOUND COUNTING

Robert W. Bistline

William H. Tyree

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A Comparison of Low-Energy Photon Detectors for Plutonium and Americium Wound Counting.

Robert W. Bistline and William H. Tyree

Abstract. A lithium-drifted silicon, Si(Li), detector system was used to predict the nuclide content and apparent depth of plutonium and americium embedded in an actual wound.

Data comparing the L x-ray energy resolution capability of a gas proportional and Si(Li) detector for a plutonium source were obtained.

A Si(Li) detector system was used with an americium source to obtain L x-ray and low-energy gamma ray absorption data with untempered Masonite absorbers. The ratios of photon absorption obtained from the Si(Li) detector system allowed apparent depth determinations to be made.

INTRODUCTION

The small crystal scintillator, NaI(Tl), has been used for some time[1,2,3] for the detection of plutonium and americium L x-rays from contaminated wounds. This detector incorporates characteristics which are still desirable for practical applications of wound counting. However, the scintillator system becomes inadequate when information about the depth, nuclide composition, or accurate quantity of the contaminant, is needed. The basic resolution capability of the sintillator system does not allow the individual lines of the L x-ray spectrum to be resolved.

Proportional detectors have been built which have much better resolution than scintillator systems.[4,5,6,7,8] These systems provided an improvement in resolution but with much lower efficiency. The utilization of dense gases[9] produced higher efficiency but with an attendant increase in background. Our experience has indicated that large proportional detectors are necessary to compensate for the limitations of gas proportional detectors and are difficult to maintain in routine operation.

Semiconductor detectors[10,11,12,13,14] show marked improvement in efficiency and resolution over the proportional detector.

The desirable features of small size, low background, high efficiencies at low energies, and excellent resolution make the Si(Li) detector useful for investigating contaminated wounds. This study showed that the Si(Li) detector system had the capability of indicating an apparent depth of material deposited in a wound and of identifying the nuclides present.

RESULTS

Using a plutonium source, a 500-mm^2 proportional detector, 5-cm deep, filled with P_{10} (90% argon and 10% methane) was used to compare the resolution of a gas proportional detector with that of an 80-mm^2 Si(Li) detector having a 3-mm depletion depth. The proportional and silicon detector spectra for the same L x-ray spectrum of plutonium are shown in Figure 1.

The Si(Li) detector system was used to obtain a series of L x-ray spectra using a 0.2 μg ^{241}Am source. Increasing thicknesses of Masonite absorbers were placed within the space between the source and the detector to obtain the series shown in Figure 2.

Transmission ratios for the silicon detector are shown in Figure 3 for untempered Masonite absorbers. The data includes ratios for the 26.36 keV-gamma ray of americium with the lower energy L x-rays. A particular energy peak indicated in a ratio represents the integration of the detected events at that peak energy.

DISCUSSION

The resolution capability of the Si(Li) detector system provides more information than can be obtained with the present scintillator system. The use of characteristic emissions from the radioisotope makes it possible to remove the influence of one isotope from the spectrum observed from a combination of two or more. This is commonly known as spectrum stripping.

The relative abundance of 13-, 17-, and 20-keV L x-rays are approximately the same for plutonium and americium as found by our measurements and measurements by other authors.[15,16,17] More precise measurements of these ratios for isotopes of plutonium and americium should be made and would be helpful for additional accuracy before using these procedures routinely.

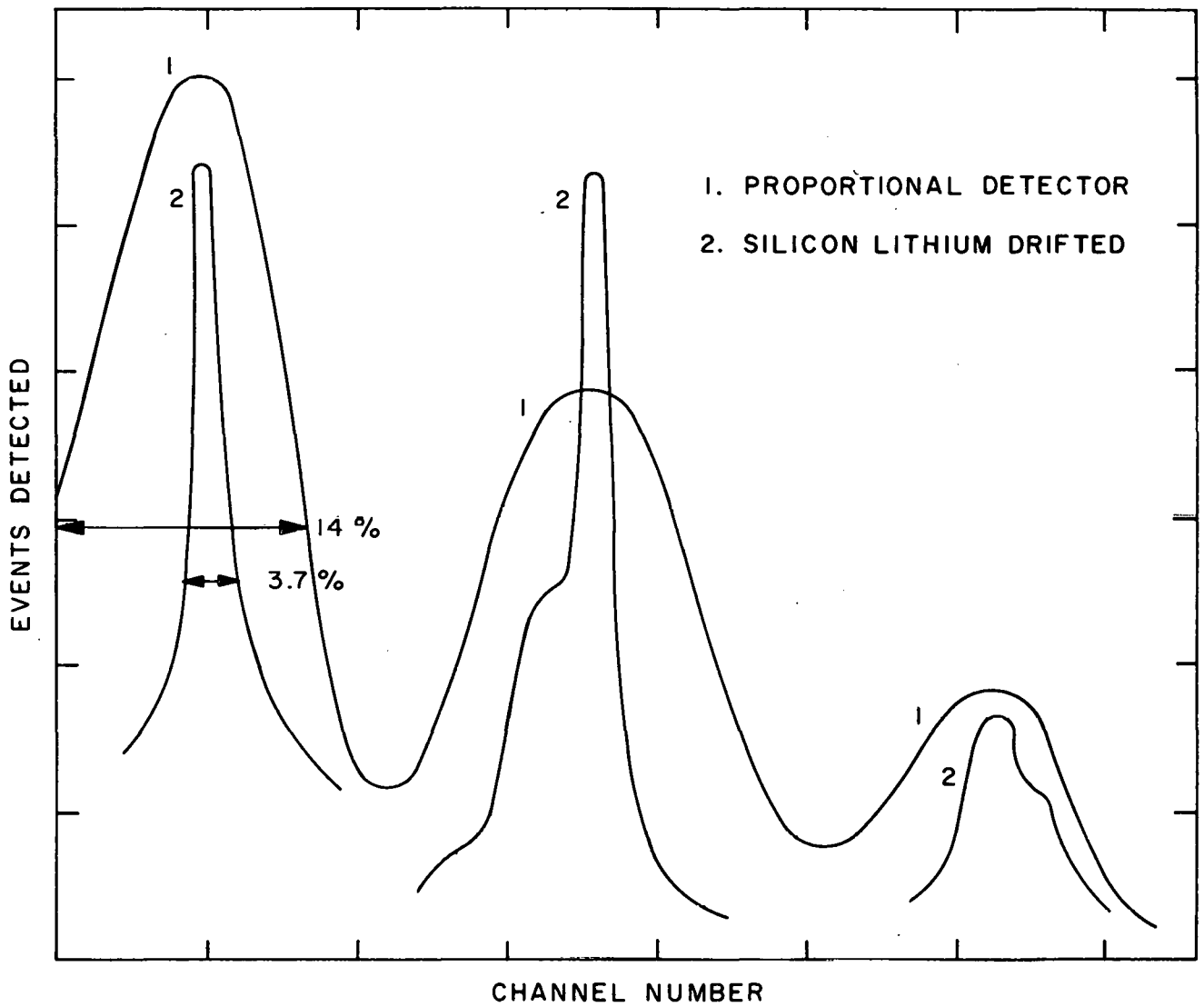


FIGURE 1. Proportional and Si(Li) Detector Spectra for the L X-ray Spectrum of Plutonium.

The data obtained from the absorption measurements with americium were utilized to determine the apparent depth of the material in an actual wound. This was achieved by selecting the absorption spectrum from the set of calibration spectra that best approximated the wound spectrum. The subtraction of the test spectrum from the wound spectrum produced the net spectrum of plutonium. From Figure 3, ratios of the various peaks gave an apparent depth of 7.9 mm.

Two methods were actually used to determine the nuclide composition of the material in a wound. The first method was a mathematical construction of the plutonium portion of the wound spectrum. An ^{241}Am spectrum which had undergone approximately the same absorption as the wound material, was selected from Figure 2 for subtraction from the wound spectrum. A ratio of the areas under each of the three major L x-ray peaks to the area under the 26.36-keV gamma ray peak was obtained from Figure 3. The ratio values gave the proper ratio of detected events to subtract from the separate peaks of the analyzed wound spectrum to obtain a plutonium spectrum. Figure 4 shows a plutonium spectrum constructed by this mathematical method. Knowing the total activity present in any one of the x-ray peaks, the ^{241}Am content of the wound can be calculated using the previously determined ratios. In addition, ratios of peak areas determined above will give apparent depth of the material with the use of Figure 3.

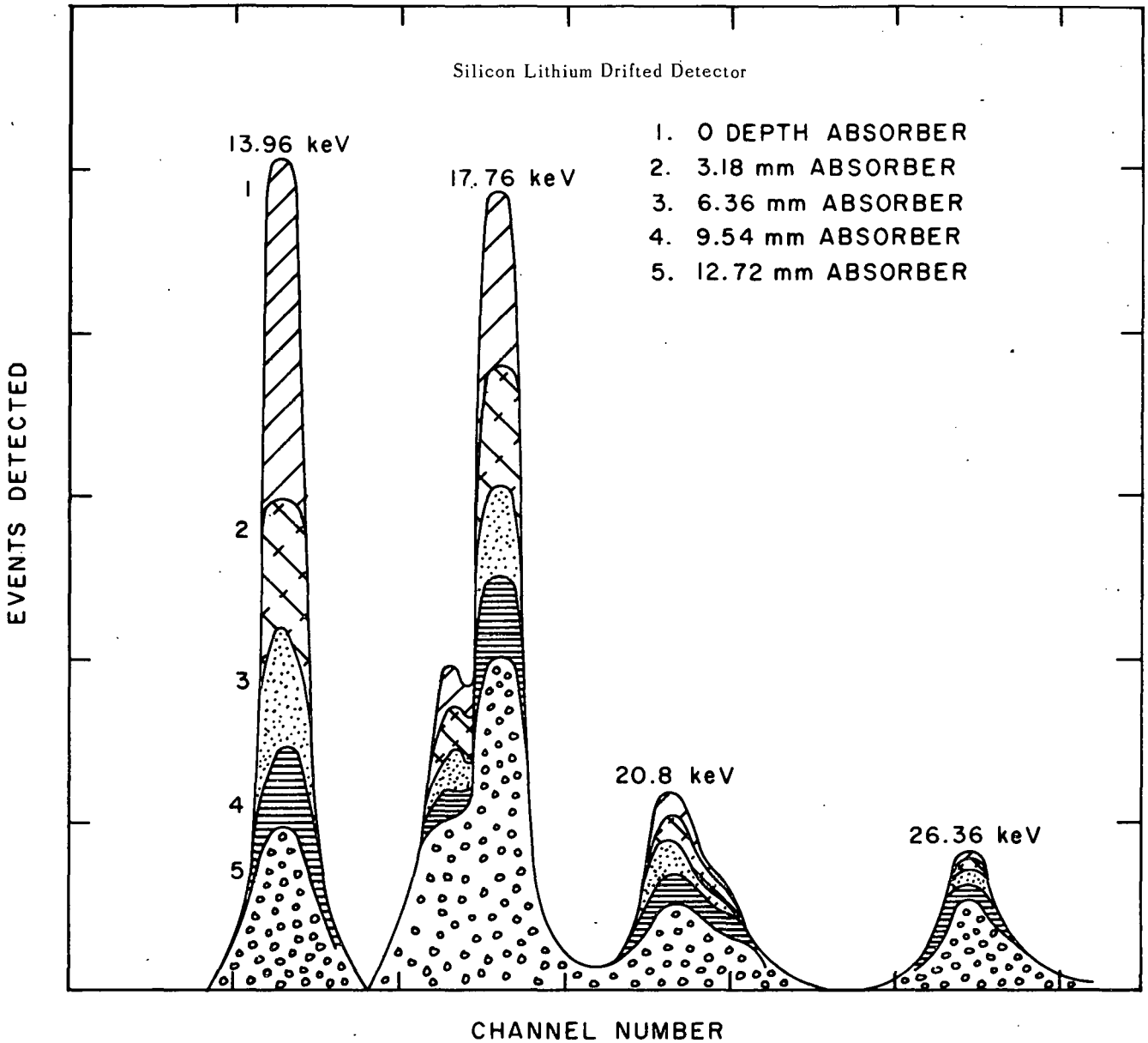


FIGURE 2. L X-ray and Low Energy Gamma Ray Spectra of Americium with Varying Thicknesses of Untempered Masonite.

The second method used the selected ^{241}Am spectrum which was then subtracted from the wound spectrum through the use of a computer type multichannel pulse height analyzer. Fractional amounts of this total spectrum were subtracted from the wound spectrum until the events recorded at the position representing the 26.36-keV gamma ray of ^{241}Am were removed. The spectrum obtained by this method is shown in Figure 5. Knowledge of the fractional portion of area subtracted from any of the three major L x-ray peaks allowed the calculation of the amounts of ^{241}Am and plutonium in the spectrum. The nearly identical spectra obtained by the two above methods reinforces the validity and usefulness of spectrum stripping with a computing type pulse height analyzer.

The analysis of a wound spectrum using either of the described procedures may be capable of giving clues to differences in translocation rates of americium and plutonium from these sites.

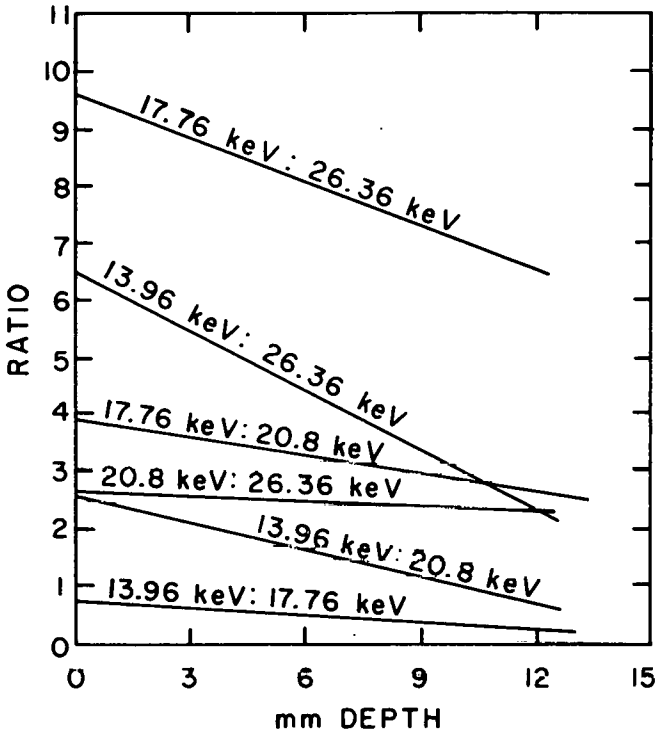
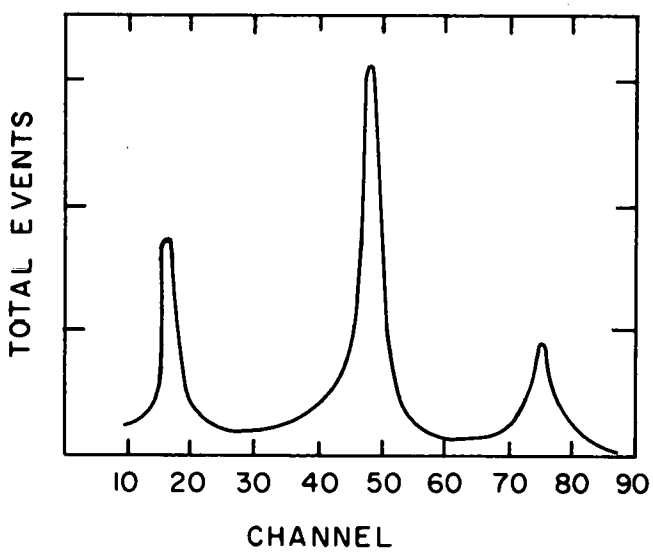


FIGURE 3. Photon Transmission ratios through untempered Masonite Absorbers as Detected by the Si(Li) detector.

The preceding procedures were used to analyze a 2-year-old wound having an originally known amount of ^{241}Am. Comparisons of the ratios of all the peaks before and after ^{241}Am stripping to those ratios in Figure 3 showed almost perfect agreement for an apparent deposition depth of 7.9 mm. The amount of ^{241}Am found by the above procedures showed very close agreement to that given by the laboratory analysis obtained from the growth corrected original material. The amount of ^{241}Am obtained by ratios from the present wound spectrum was about 10% less, which may indicate a more rapid loss of ^{241}Am than plutonium from the wound site. An additional source of error not considered in this procedure was the self-absorption of low-energy photons dependent upon particle size. Correction of the data for self-absorption would make an even greater difference in the determined amounts of ^{241}Am.

FIGURE 4. Net Plutonium Spectrum from Point-by-Point Subtraction of ^{241}Am Ratios as Shown in Figure 3.



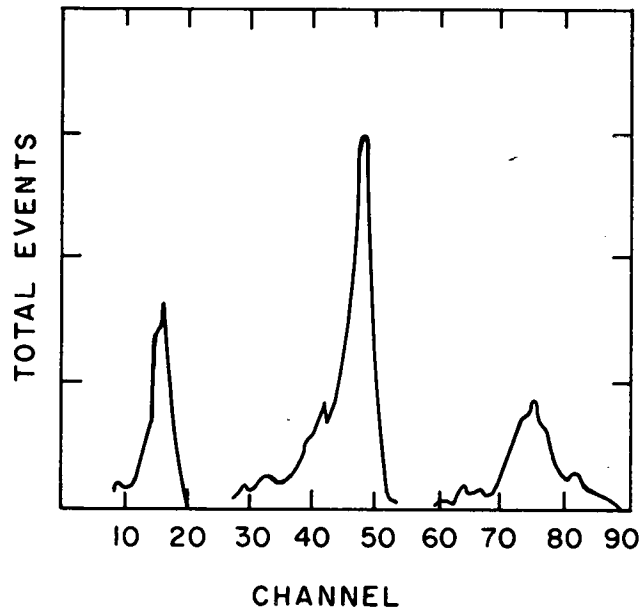


FIGURE 5. Net Plutonium Spectrum from the Use of the Computer Function of a Multichannel Pulse Height Analyzer.

SUMMARY

A proportional detector produced 14 percent resolution at the 13.6-keV L x-ray line of plutonium. A Si(Li) detection system produced 3.7 percent resolution for the same source and energy. Using an unattenuated americium source, a ratio of 3.9 for the absorption of the 17.76-keV compared to the 20.8-keV photon energy was observed with an 80-mm^2 Si(Li) detector having a 3-mm depletion depth. Using the same detector and 17.2 keV and 20.2 keV the same ratio was obtained for a plutonium source.

Discrete absorption data, which were obtained for americium, provided a means of estimating depths for actual wounds. The Si(Li) detector system used on a wound containing a composite of americium and plutonium indicated an apparent depth of material of 7.9 mm.

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