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A Si(Li)-NaI(Tl) DETECTOR FOR DIRECT MEASUREMENT
OF PLUTONIUM IN VIVO

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I. Introduction

Measurements of Pu in human lungs *in vivo* is a difficult problem but, when in addition to the Pu, ^{241}Am is also present, this problem becomes formidable. At present there are no adequate detectors with which one can measure Pu in the presence of ^{241}Am directly. The problem is usually circumvented by measuring Pu indirectly, using the ^{241}Am as a tracer. This technique is based on *a priori* knowledge of the ratio of the ^{241}Am activity to that of Pu *in vivo*. However, this activity ratio *in vivo* may not be the same as that in the contaminant especially if there is more than one contaminant. Consequently, such an indirect measurement lacks the reliability of a method whereby the Pu activity is observed directly.

At the Los Alamos Workshop on Low Level Transuranic Measurements¹ we discussed the potential of a Si(Li)-NaI(Tl) detector system for measuring the UL x rays produced in the decay of Pu. In this paper we describe the conceptual design and expected performance of such a system. The detector can resolve the UL x rays from the NpL x ray thus permitting direct measurement of Pu in the presence of ^{241}Am . The expected performance of the system was determined from measurements of lung phantoms using a prototype Si detector.

II. Si(Li)-NaI(Tl) Detector

The detector system currently being developed (Fig. 1) consists of an array of six Si(Li) detectors in a liquid nitrogen (LN) cooled cryostat surrounded by a large NaI(Tl) anticoincidence scintillator for background suppression. Each Si(Li) detector, 34 mm dia. x 7 mm thick, is mounted on its edge and has a thin window on its entrance and exit surface. A one-piece Be entrance window on the cryostat which extends beyond the entire detector array minimizes losses of x rays incident at oblique angles. This detector configuration, which is similar to the one developed for measurement of radionuclides in soil,² combines the salient features of the phoswich detector with the superior performance of semiconductor detectors:

- High energy resolution which permits separation of the UL x rays due to Pu from the NpL x rays due to ^{241}Am and which provides for a narrow energy window about the peak for background reduction.
- High intrinsic x-ray detection efficiency equal to that of the phoswich.
- Active background suppression similar to that of the phoswich.
- Low sensitivity to γ -ray background because of the low Z of Si.

The projected area of the Si(Li) detector array is approximately 50 cm^2 . While this area is less than half that of a 5 inch diameter phoswich it does not necessarily indicate a proportionate decrease in signal detection. A study by Newton et al³ suggests that the standard 5 inch phoswich may well be larger than optimum for best signal-to-background ratio. It also points out that judicious placement of individual detectors with respect to the anatomical features may result in improved signal-to-background ratio. The optimum configuration of the detector array will be determined from investigations now being conducted using the LLNL^{*} phantom as a model.

III. Comparison of X-Ray Detector Characteristics

In order to measure Pu in the presence of ^{241}Am the UL_{β_1} x ray (17.22 keV) should be resolved from the NpL_{β_1} x ray (17.75 keV). This requires a detector with a FWHM resolution of 0.5 keV or better. Table I lists the calculated intrinsic line widths and the observed resolutions for several detectors used in measurements of Pu in lungs. The second column (ϵ) gives the energy that must be expended in the detector to create one photoelectron or an electron pair and the third column (F) is the Fano factor. The calculated line width is that of a 17.2 keV γ ray, not of the x ray. The

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observed FWHM of UL and NpL x rays may be several percent larger than that of γ rays due to the tails in the Lorentzian distribution.⁴ The measured detector resolution (column 4) is considerably wider than the intrinsic line width, as expected.

Only Si and Ge detectors have the resolution required to resolve the UL x rays from the NpL x rays. For a given diameter, a somewhat thicker Ge detector gave the same FWHM as a Si detector. This is evident in Table I in which the thickness of the Ge detector is 10 mm and that of the Si is 7 mm. A thicker Ge detector is necessarily more sensitive to γ -ray background yet not any more efficient for the detection of x rays.

In addition to being a function of thickness the detector background sensitivity is also a function of the atomic number Z of the detecting medium. The Z for several detectors is given in Table II. Si has the lowest Z and hence is the least sensitive to γ ray background for a given thickness.

When γ rays from ^{241}Am induce characteristic K x rays in the detector and the latter escape, a satellite escape peak of energy $E_{\gamma} - E_{\text{x}}$ is formed. Such peaks are undesirable when they interfere with the peaks of interest. In the case of NaI(Tl), for example, the iodine K_{α} escape peak, due to the 59.5 keV γ ray from ^{241}Am , interferes with the UL x rays as shown in the following section. In the case of Ge the K_{α} escape peak, due to the 26.3 keV γ ray in ^{241}Am , interferes with the UL_{β} peak complex. In Si the escape probability is very small due to the low energy of the K_{α} x ray.

IV. Performance of Si(Li)-NaI(Tl) Detector vs. Phoswich Detector

The expected performance of the new Si(Li)-NaI(Tl) system relative to a phoswich was determined with a prototype detector consisting of a single Si(Li) detector and a 5"x4" NaI(Tl) scintillator. The prototype Si(Li) detectors were 32-34 mm in diameter and 6-7 mm thick, as in the projected array. The 4" dia. phoswich,* consists of a front 1 mm thick NaI(Tl) crystal

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mounted on a 2" thick CsI(Na) crystal. The superior energy resolution of the Si(Li) detector relative to the phoswich is clearly evident in the ^{239}Pu spectra in Figs. 2 and 3. In Fig. 2 the UL_β and UL_γ peaks are completely separated and well-defined, even with the 5 cm absorber to simulate the lungs plus chest wall, whereas in Fig. 3 they form a single broad composite UL x-ray peak. The total energy window required with the Si(Li) detector to measure the UL x rays is 1/5 that required with the phoswich. The measured ambient plus ^{40}K background per keV is comparable on a unit area basis for the two detectors. Therefore, the background in the window, for a unit area Si(Li) detector, is 5 times lower.

Measurements with a phoswich of Pu in the presence of ^{241}Am present considerable problems. Since it cannot resolve the UL_β from the UL_γ peaks as shown in Fig. 3 it certainly cannot resolve the NpL x-ray peaks resulting from the decay of ^{241}Am from the UL x-ray peaks resulting in the decay of Pu. In Fig. 4, which shows a superposition of UL and NpL x-ray spectra, these peaks are resolved with the Si(Li) detector with absorber present.

A more significant problem than is generally recognized is illustrated in Fig. 5. The NpL x rays are highly absorbed with 5 cm of tissue-equivalent absorber and are quite obscured by the tail of the iodine K escape peak associated with the 59.5 keV γ ray. Degradation of this γ ray, due to scatter in tissue, accounts for the low-energy spread of this peak. The tail of the 26.3 keV peak, which is highly absorbed and masked by the escape peak, also falls into the phoswich energy window. With the Si(Li) detector the 26.3 keV peak is completely outside the x-ray energy window (Fig. 2).

The above ^{241}Am γ -ray contributions to the phoswich window, together with a continuum of the scattered ^{241}Am γ rays, are several times as large as that from the unresolved NpL x rays. As a result of all these contributions (x and γ rays), for Pu with 0.1% ^{241}Am (weapons grade) there are three times as many counts in the window due to the ^{241}Am as there are from the UL x rays. In comparison, the corresponding count ratio for the Si(Li) system is only about 0.4. Most of the counts in the Si(Li) energy windows are from the NpL x-ray peaks, which are not fully resolved, as seen in Fig. 4.

The capability of the Si(Li) detector to separate the UL x rays from those produced in the decay of other actinides is demonstrated in Fig. 6. The figure shows the spectra of the L_{β} x rays from four actinide sources: PaL_{β_1} from ^{237}Np , UL_{β_1} and UL_{β_2} from ^{239}Pu , NpL_{β_1} and NpL_{β_2} from ^{241}Am , and PuL_{β_1} and PuL_{β_2} from ^{244}Cm .

V. Minimum Detectable Pu Activity

A Si(Li) prototype detector 34 mm diameter was used to simulate two six-detector arrays in a lung phantom measurement. The experimental arrangement inside the radiation shield is shown in Fig. 7. The Si(Li) detector is mounted within the paddle-shaped section (square) on the top end of the cryostat. The cryostat extends through the shield floor where the liquid nitrogen dewar (not shown) is located. The detector is mounted on its edge and has a thin window on each side. A 5"x4" NaI(Tl) anticoincidence scintillator is on the left and a 2.5 cm thick tissue-equivalent absorber (Rando), to simulate the chest wall, with two metal strips (not shown), to simulate the ribs, are on the right. The lung phantom is next to the absorber. Two different lung phantoms were employed in order to observe the effect of ^{241}Am on Pu measurements. One of the lung phantoms (obtained from a LLNL phantom) was loaded with ^{239}Pu and the other with ^{241}Am . A sensitivity of 0.03 cpm/nCi ^{239}Pu was obtained in this simulation.

Results from the measurements with the lung phantoms and background measurements, were combined to produce simulated spectra for lung burdens as low as 15 nCi ^{239}Pu . For each lung burden, one spectrum was obtained where ^{241}Am was absent and another where it was present. The spectrum for a lung burden of 30 nCi is shown in Fig. 8 where each channel is equivalent to 60 eV. Although the number of counts per channel are low, the UL_{β} and UL_{γ} x-ray peaks are quite discernable. In the lower spectrum ^{241}Am is absent while in the upper spectrum there is 0.1% ^{241}Am .

The ^{241}Am 59.5 keV and 26.3 keV γ -ray peaks in the upper spectrum are clearly visible while the NpL x-ray peaks are too attenuated to be discerned. The total 2.2 keV measurement interval is determined by two separate windows, one about each of the UL_{β} and UL_{γ} x-ray peaks. In that interval there are 42

counts from the UL x rays and 11 counts from ^{241}Am , most of which are due to the NpL x rays. There are 35 background counts, the average background rate being close to 1 cpm which includes the ^{40}K subject background.

Averaged ratios obtained for the total $\text{UL}_{\beta_{1,3,5}}$ and UL_{γ} x-ray counts to the square-root-of-the-background for the 2000 sec simulated lung burden measurements are shown in Table III. These ratios are close to those expected on the basis of the activity and therefore are a measure of the confidence in the detection of the activity. The activity measurement accuracy for a single sample measurement is shown in the last column of the table. The accuracy applies to the assay rather than to the detection of the activity. The background used in the calculations includes the ambient background, that from ^{40}K in the subject, and the effective background contribution due to ^{241}Am when it is present. The values obtained for ^{239}Pu lung burdens of 15 and 30 nCi are shown first without and then with 0.1% ^{241}Am . The confidence in the presence of true activity,* corresponding to the ratio of 2.5 for 15 nCi of ^{239}Pu alone is nearly 90%. With 0.1% ^{241}Am this ratio becomes 2.1 and the confidence is 85%. For the 30 nCi cases the confidence in either case is at least 95%. The reduction in the ratios and activity measurement accuracies caused by the presence of 0.1% ^{241}Am is only ~20%.

The effective value for the square root of the background, where ^{241}Am is present, includes the statistical uncertainty due to the ^{241}Am counts in the energy window, which consist of unresolved NpL x rays and scattered γ rays (26.3 keV and 59.5 keV). The number of unresolved NpL x rays in the window is inferred from the intensity of the measured 26.3 keV peak.

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The risks of concluding that activity is present when there is none (false positive) or absent when there is some (false negative) are considered equal.

VI. Concluding Discussion

The results of the measurement using the prototype Si(Li) detector with a NaI(Tl) anticoincidence scintillator to simulate a pair of six-detector arrays indicates that, even with 0.1% ^{241}Am (weapons grade) present, the confidence for correctly determining the presence of 15 nCi ^{239}Pu is 85%. This is the confidence corresponding to a peak to square-root-of-background ratio of 2.1 (Table III). It follows that for 85% confidence the minimum detectable activity (MDA) is 15 nCi. Increasing the counting time from 2000 sec to 4000 sec increases the confidence for the correct detection of this activity to nearly 95%.

Equivalent values for MDA of ^{239}Pu (^{241}Am absent) obtained with a pair of phoswich detectors by Anderson⁷ and by Newton et al³ are approximately 20 and 30 nCi, respectively. While improved detector geometry may lower the MDA for the phoswich,³ the Si(Li) system promises at least as good a sensitivity in the presence of ^{241}Am as that of the phoswich. However, in the phoswich case the sensitivity is necessarily obtained in the absence of ^{241}Am .

The performance of Ge detectors in the measurement of Pu in lungs via the UL x rays has been investigated by Falk et al,⁸ Newton, et al,³ and Berger and Goans.⁹ Although the energy resolution of the Ge detector could be as high as that of the Si(Li) detector, the peak-to-background ratio would not be as high. The greater thickness and higher Z of Ge detectors inherently results in a higher γ -ray background. Moreover, the above Ge detector configurations do not permit anticoincidence background suppression.

The importance of background suppression is demonstrated in Fig. 8, in which the UL x-ray peaks due to 30 nCi of ^{239}Pu are visible even in the presence of ^{241}Am . Had the background been several times higher, as it would have been without the NaI(Tl) anticoincidence, the ability to discern the peaks would have been seriously impaired, notwithstanding the high resolution of the Si(Li) detectors. This has been substantiated by Newton et al,³ who observed that, although Ge detectors have high resolution, without substantial reduction in background their sensitivity will not compare favorably with that of a phoswich. The design of the present detector system

using Si(Li) detectors, with their inherently lower γ -ray sensitivity, and anticoincidence background suppression combines the salient features of the phoswich with the high energy resolution of semiconductor detectors. Furthermore, the high energy resolution allows the separation of the UL x rays from the NpL x rays thus permitting the measurement of Pu in the presence of ^{241}Am -- a capability that did not appear promising at low levels with an array of Ge detectors.¹⁰

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TABLE I

X-RAY ENERGY RESOLUTION OF DETECTORS
USED IN MEASUREMENTS OF Pu IN LUNGS

DETECTOR	ϵ	F	CALCULATED ^(a) INTRINSIC FWHM FOR 17.2 keV γ RAY (keV)	OBSERVED FWHM FOR 17.8 keV NpL $\beta_{1,3,5}$ X RAY (keV)
NaI(Tl) ^(b)	300 eV/pe	1	5.4	9.0 ^(f)
GSPC(Xe) ^(c)	21.9 eV/e-i	0.13	0.52	1.0
Ge ^(d)	2.98 eV/e-h	0.13	0.19	0.4
Si ^(e)	3.81 eV/e-h	0.13	0.22	0.4

^(a) $FWHM = 2.355 \sqrt{F \cdot \epsilon \cdot E}$, where $E = 17.2$ keV

^(b) 10 cm dia. (Harshaw phoswich)

^(c) 20 cm dia. (D. F. Anderson⁴, H. E. Palmer⁵)

^(d) 3.4 cm dia. x 10 mm thick (ORTEC HPGe LEPS)

^(e) 3.4 cm dia. x 7 mm thick [LBL-ANL edge-mounted Si(Li)]

^(f) FWHM is of composite peak (NpL $_{\alpha}$, NpL $_{\beta}$, NpL $_{\gamma}$)

TABLE II

CHARACTERISTICS OF DETECTORS USED
IN MEASUREMENTS OF Pu IN LUNGS

DETECTOR	Z	K_{α} X-RAY ESCAPE (keV)
NaI(Tl)	53	28.6
GSPC(Xe)	54	29.8
Ge	32	9.9
Si	14	1.7*

*Chance of escape is negligible.

TABLE III

SIMULATED 2000 SEC LUNG BURDEN MEASUREMENT

LUNG BURDEN		CONFIDENCE IN PRESENCE OF TRUE ACTIVITY peak ^(a) / $\sqrt{\text{bkg}^{(b)}}$	MEASUREMENT ACCURACY ($\Delta A/A$)
²³⁹ Pu (nCi)	²⁴¹ Am (% ²³⁹ Pu mass)		
15	---	2.5	0.47
15	0.1	2.1	0.55
30	---	5.1	0.27
30	0.1	3.8	0.33

(a) Sum of UL _{β} and UL _{γ} x-ray counts.

(b) Includes ambient, ⁴⁰K, and effective background due to ²⁴¹Am when present.

Captions

- Fig. 1. Configuration of proposed Si(Li)-NaI(Tl) lung monitor. An array of edge-mounted Si(Li) detectors having thin entrance and exit windows are operated in anticoincidence with large NaI(Tl) scintillator for background suppression.
- Fig. 2. X and γ -ray spectrum with prototype Si(Li)-NaI(Tl) detector from ^{239}Pu source containing trace ^{241}Am impurity. Absorber simulates lungs and chest wall. UL_β and UL_γ peaks are separated and well-defined. The combined width of the two energy windows used is 2.2 keV.
- Fig. 3. X and γ -ray spectrum with phoswich from ^{239}Pu source containing trace ^{241}Am impurity. Absorber simulates lungs and chest wall. UL_β and UL_γ x rays produce single broad peak. The width of the energy window used is 11 keV.
- Fig. 4. Superposition of UL and NpL x-ray spectra with Si(Li) detector from ^{239}Pu and ^{241}Am sources. Although x-ray energies are spread by absorber, principal UL_β and UL_γ peaks are well resolved from the corresponding NpL_β and NpL_γ peaks. The broad energy window used with phoswich is shown for reference on the bottom.
- Fig. 5. Spectrum obtained with phoswich showing iodine K x-ray escape peak associated with 59.5 keV γ ray from ^{241}Am . Low-energy spread due to tissue-scatter of γ ray further extends escape peak tail into UL x-ray window. NpL x rays and 26.3 keV γ rays from ^{241}Am are highly absorbed and therefore obscured by escape peak.
- Fig. 6. Spectra showing capability of Si(Li) detector to separate UL x rays from those produced in the decay of other actinides. L_β x-ray spectra are from four actinide sources: PaL_{β_1} from ^{237}Np , UL_{β_1} and UL_{β_2} from ^{239}Pu , NpL_{β_1} and NpL_{β_2} from ^{241}Am , and PuL_{β_1} and PuL_{β_2} from ^{244}Cm . Source strengths are relative to that of ^{239}Pu .

Fig 7. Si(Li)-NaI(Tl) prototype detector and lung phantom shown inside radiation shield. Si(Li) detector is within paddle-shaped endcap (square) of the cryostat, which is against the scintillator. Rando absorber is sandwiched between the endcap and the lung phantom on the right.

Fig. 8. Spectra from ^{239}Pu and ^{241}Am in the lung phantoms produced in simulation of two six-detector arrays with prototype Si(Li) detector and NaI(Tl) anticoincidence scintillator. The lung burden for both spectra is 30 nCi ^{239}Pu . In addition to Pu the upper spectrum also contains 0.1% ^{241}Am (weapons grade). The UL_β and UL_γ x-ray peaks are apparent in both spectra while the NpL x rays are too attenuated to be seen. The 59.5 keV and 26.3 keV ^{241}Am γ -ray peaks are clearly visible.

Fig. 1

Fig. 2

Fig. 3

3 cm TISSUE-EQUIVALENT ABSORBER (RANDO)

Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8