L X-RAY SPECTROMETRY IN VIVO WITH A Si(Li)-NaI(Tl) DETECTOR*

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ABSTRACT

A new Si(Li)-NaI(Tl) spectrometer was developed for measurements of UL x rays from $^{239}$Pu in lungs of exposed persons. The spectrometer consists of six large, cooled Si(Li) detectors mounted on edge to provide two windows, one facing the lungs and the other a NaI(Tl) scintillator for anticoincidence background suppression. The sensitive area of the array is $54.5 \text{ cm}^2$ and the FWHM resolution at $17 \text{ keV}$ is $190 \text{ eV}$. The ambient background count rate per minute in the energy band of interest of the Si(Li)-NaI(Tl) detector is $0.13$. This is $<1/4$ of that of an equal-area HPGe detector and $1/15$ of an equal-area phoswich. The sensitivity of this detector for $^{239}$Pu approaches the maximum achievable, where the minimum detectable level is fundamentally limited by the lung geometry (signal) and the natural radioactivity in the human body (background). With this detector one can measure low level Pu in human lungs directly even in the presence of $^{241}$Am. A spectrum of ZrK x rays from $^{92}$Nb (simulating Pu), obtained from lungs of a human subject in vivo, demonstrates the detector performance.

I. INTRODUCTION

Prior to the development reported here low levels of plutonium in human lungs could not be measured unambiguously. At present, the Pu level is usually inferred from a measurement of $^{241}$Am which is used as a tracer. Such an indirect measurement has many uncertainties and lacks the reliability of a direct measurement[1]. In 1984 we reported[2] the feasibility of a Si(Li)-NaI(Tl) detector system for measuring Pu in human lungs directly by observing the UL x rays emitted in the Pu decay. Experiments in which a prototype detector was used with phantom lungs and tissue-equivalent absorbers showed that an array of Si(Li) detectors surrounded by a large NaI(Tl) scintillator for background suppression, could be suitable for direct measurements of Pu in the presence of $^{241}$Am. Such a detector array would have to have a FWHM resolution at $17 \text{ keV}$ of less than $500 \text{ eV}$ and be large enough to measure the minimum detectable level in a reasonable time[3]. The minimum detectable activity is fundamentally limited by the signal strength as determined by the lung geometry and by the background, both the ambient and the natural radioactivity of the human body.

In this paper we describe an array of cooled large area Si(Li) detectors with anticoincidence background suppression. Performance data are given and results of the initial measurements of human subjects in vivo are presented.

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

The detector system consists of a rectangular array of six planar Si(Li) detectors mounted in a paddle-shaped, liquid-nitrogen (LN) cooled cryostat that is surrounded on three sides by a large NaI(Tl) scintillator (Fig. 1). The scintillator, $30 \text{ cm}$ in diameter by $10 \text{ cm}$ thick, is operated in anticoincidence with the Si(Li) detectors to suppress y-ray background arising from Compton interac-

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Fig. 1 Configuration of the Si(Li)-NaI(Tl) detector. An array of six edge-mounted Si(Li) detectors having entrance and exit windows is operated in anticoincidence with a large NaI(Tl) scintillator for background suppression.

Each Si(Li) detector is 34 mm in diameter by 7 mm thick giving a total sensitive area of 54.5 cm². The Si(Li) crystals are mounted on edge and have windows on the entrance and exit surfaces. The cryostat has separate entrance windows for each detector, made of 0.25 mm thick Be, and a common exit window made of 1.65 mm thick Al. A photograph of the detector system, taken during its assembly, is shown in Fig. 2. The Be windows of the detector array are seen in the mirror below the cryostat paddle. The NaI(Tl) scintillator is attached to the Si(Li) detector array through its flange, thereby accommodating the paddle inside the 25 mm deep recess against the 0.25 mm thick Al window on the NaI(Tl). The detector assembly is mounted on a motor-driven gantry with X, Y and Z translation and tilt provisions with respect to the vertical of +/-30°. The detector assembly is housed inside a 2x2x2 m³ radiation shield and is equipped with an automatic LN-fill system.

In the present configuration, shown in Figs. 1 and 2, all the detectors are mounted in a single paddle-shaped cryostat rather than in individual cryostats. A power or vacuum failure could result in catastrophic damage to the entire detector array. To guard against such occurrences we designed a monitoring system which senses the AC and DC voltages, vacuum ion-pump current and liquid-nitrogen overflow. The Argonne site-wide computer surveillance system monitors the state of these
sensors and provides prompt notification in the event of failure. Over the past three years we have had several failures none of which were catastrophic or even deleterious to the performance of the detectors.

In order to measure Pu in the presence of $^{241}$Am, the UL$_{\delta 1}$ and UL$_{\gamma 1}$ x-ray peaks due to Pu must be resolved from the NpL$_{\delta 1}$ and NpL$_{\gamma 1}$ x-rays due to $^{241}$Am. Thus, energy resolution, consistent with high efficiency and low background, was the guiding principle in the design of this system. The choice of Si over Ge for this type of detector application has been thoroughly examined in an earlier study[2]. The lower Z of Si and hence lower sensitivity to γ-ray background was the primary reason for its choice. The detector dimensions were selected primarily on the basis of resolution considerations. The design of this type of detector array has been reported in detail in an earlier publication[4].

III. DETECTOR PERFORMANCE

The resolution of the 6-detector array was measured with a $^{241}$Am source. For the NpL$_{\delta 1}$ peak complex at 17.75 keV, FWHM=390 eV, and for the γ-ray peak at 59.5 keV, FWHM=480 eV. The ability of this spectrometer to resolve the UL x rays from the NpL x-rays is illustrated by the spectrum in Fig. 3. This spectrum was acquired in a 30-min count of simulated lungs containing 37 kBq (1000 nCi) $^{239}$Pu and 2.0 kBq (55 nCi) $^{241}$Am (0.1% by weight). The lungs were inserted in a phantom thorax having a muscle equivalent chest-wall thickness of 23 mm. This phantom thorax would represent, for example, that of a 1.78-m (70 in) tall, middle-aged male worker weighing 73 kg (160 lb)[5]. It is shown in the next section that by selecting narrow energy bands containing most of the counts in the UL x-ray peaks of interest, the interference from the NpL x-ray peaks is insignificant, thus providing the capability of measuring Pu in the presence of $^{241}$Am.

The sensitivity of this detector system to background radiation as compared with that of other detectors is given in Table I. The count rate shown is the ambient background inside the radiation shield in the UL x-ray energy range. The listed count rates of all the detectors are given per unit detector area, where the unit is the 54.5-cm$^2$ area of the Si(Li) array. The actual dimensions of the HPGe planar detector* are: diameter=25 mm, thickness=10 mm (at 17.75 keV, FWHM=410 eV). The background count rate in the Si(Li) detector array is seen to double without anticoincidence background suppression. The background count rate in the HPGe detector is more than twice that in the Si(Li) detector alone, and more than 4 times that in the Si(Li) detector when it is used with anticoincidence. The background count rate in the phoswich per unit energy is only ~40% higher than that in the Si(Li)-NaI(Tl) system, but due to the poor resolution of the phoswich its background is 15 times higher in the x-ray energy range of interest. It is shown below that the ambient background count rate of 0.13 min$^{-1}$ keV$^{-1}$ in the present Si(Li)-

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TABLE I
AMBIENT BACKGROUND COUNT-RATE* IN THE REGIONS OF INTEREST (ROI) FOR MEASUREMENT OF UL X-RAYS

<table>
<thead>
<tr>
<th>Detector</th>
<th>Count-rate per unit of energy in ROI† (min⁻¹ keV⁻¹)</th>
<th>Width of ROI† (keV)</th>
<th>Count-rate in ROI (min⁻¹)</th>
<th>Relative</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si(Li)-NaI(Tl)</td>
<td>0.13</td>
<td>1</td>
<td>0.13</td>
<td>1</td>
</tr>
<tr>
<td>Si(Li)</td>
<td>0.26</td>
<td>1</td>
<td>0.26</td>
<td>2</td>
</tr>
<tr>
<td>HPGe</td>
<td>0.55</td>
<td>1</td>
<td>0.55</td>
<td>4.2</td>
</tr>
<tr>
<td>NaI(Tl)-CsI(Tl)</td>
<td>0.18</td>
<td>11</td>
<td>1.98</td>
<td>15</td>
</tr>
</tbody>
</table>

* Normalized to Si(Li) detector array area of 54.5 cm².
† ROIs are 17.0-17.4 keV and 19.9-20.5 keV for both the Si(Li) and HPGe detectors, and 13-24 keV for the NaI(Tl)-CsI(Tl) phosphor system.

NaI(Tl) system is significantly lower than the combined count rate from the natural radioactivity in a human subject and from the ambient background, and hence the ambient background count rate is not the principal factor limiting the minimum detectable radioactivity level.

IV. MEASUREMENTS IN VIVO

Average count rates with and without anticoincidence background suppression are shown in Table II for four unexposed male subjects measured with the Si(Li)-NaI(Tl) spectrometer positioned over the right lung. The anticoincidence is seen to reduce the background in vivo by nearly a factor of 3. If it were possible to reduce or even eliminate the ambient background count rate of 0.13 min⁻¹ keV⁻¹ (Table I), the unexposed subject count rate of 0.23 min⁻¹ keV⁻¹ (Table II), which includes the ambient background count rate, presumably would be reduced to 0.1 min⁻¹ keV⁻¹. Thus, even in the virtual absence of the ambient background radiation, the minimum detection level would be significantly limited by the natural radioactivity of the human body.

The ambient background spectrum with anticoincidence suppression is shown in Fig. 4. The ThL x-rays at 13, 16.2, and 19 keV in the background spectrum are probably due to trace contamination (<2 ppm) by natural ²³⁸U in the Be windows of the Si(Li) array. The presence of the 46.5 keV γ-ray of ²¹⁰Pb in the background spectrum is attributed to lead solder on the preamplifier components inside the cryostat paddle.
The spectrum with the detector system positioned over the right lung of a typical unexposed male subject is shown in Fig. 5. One discernible effect of the presence of the unexposed subject is to elevate the count rate of the continuum seen in the ambient background spectrum (Fig. 4).

Relationships between x-ray count rate and chest-wall thickness were determined by counting simulated lungs uniformly loaded with 239Pu in the Livermore phantom [6]. Table III gives the count rate in the two UL x-ray regions per kBq of 239Pu activity in both lungs for a muscle-equivalent chest-wall thickness of 23 mm. These two regions constitute an energy band of 980 eV that contains 80% of the total counts in the two x-ray peaks. Also shown in the table are the count rates in the two UL x-ray regions due to 241Am when it is present at 0.1% of the 239Pu mass, and the count rates for a typical unexposed male subject. The data of Table III indicate that the interference from 0.1% 241Am constitutes only 7.4% of the total x-ray count in the two UL x-ray regions. The interference due to the trace contamination

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**Table II**

<table>
<thead>
<tr>
<th>Anticoincidence Background Suppression</th>
<th>Average Count-Rate (min⁻¹ keV⁻¹)</th>
<th>Relative</th>
</tr>
</thead>
<tbody>
<tr>
<td>On</td>
<td>0.23</td>
<td>1</td>
</tr>
<tr>
<td>Off</td>
<td>0.64</td>
<td>2.8</td>
</tr>
</tbody>
</table>

*Four subjects counted, each for 60 min; x-ray region is 17-20 keV.*

Fig. 4 Spectrum of detector ambient background with anticoincidence shield. The THL x rays are due to trace contamination (< 2 ppm) of natural 238Pu in the Be windows of the cryostat containing the Si(Li) detectors. The interference of these peaks with UL x rays of interest is negligible. The 46.5 keV y-ray is attributed to 210Pb in solder.

![Fig. 4 Spectrum of detector ambient background with anticoincidence shield.](image)

![Fig. 5 Spectrum of the natural radioactivity from the right lung of a typical unexposed male subject.](image)
TABLE III
COUNT RATE FROM LUNGS FOR A TYPICAL MUSCLE-EQUIVALENT CHEST-WALL THICKNESS OF 23 mm

<table>
<thead>
<tr>
<th>Energy Region (keV)</th>
<th>X-ray</th>
<th>$^{239}$Pu ($\text{min}^{-1}\text{kBq}^{-1}$)</th>
<th>Additional Counts from $0.1%$ $^{241}$Am ($\text{min}^{-1}\text{kBq}^{-1}$)</th>
<th>Unexposed Average Male ($\text{min}^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>17.0-17.4</td>
<td>UL$\gamma_1$</td>
<td>$0.95 \times 10^{-1}$</td>
<td>$7.6 \times 10^{-3}$</td>
<td>$0.64 \times 10^{-1}$</td>
</tr>
<tr>
<td>19.9-20.5</td>
<td>UL$\gamma_1$</td>
<td>$1.05 \times 10^{-1}$</td>
<td>$8.4 \times 10^{-3}$</td>
<td>$1.83 \times 10^{-1}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$2.0 \times 10^{-1}$</td>
<td>$1.6 \times 10^{-2}$</td>
<td>$2.5 \times 10^{-1}$</td>
</tr>
</tbody>
</table>

in the Be windows is negligible. It can be seen from these data that a $^{239}$Pu lung content of 0.65 kBq (17.6 nCi) can be measured in one hour with a precision corresponding to twice the square root of the background from an unexposed subject. Our results suggest that if the number of detectors were increased to cover both lungs optimally, the $^{239}$Pu lung content that could be measured in one hour with the same statistical precision would be reduced by less than a factor of two, i.e., $0.4 \times 10^{-1}$ kBq ($10$ nCi).

In Fig. 6, the spectra of 0.65 kBq (17.6 nCi) $^{239}$Pu and 0.035 kBq (0.95 nCi) $^{241}$Am (0.1% by mass) in simulated lungs inserted in the phantom thorax (muscle-equivalent chest-wall thickness of 23 mm) are shown convoluted with a spectrum of a typical unexposed male subject. Comparison of Fig. 6 and Fig. 5 shows that the only readily discernible difference between the spectra in these two figures is the presence of the 59.5 keV $\gamma$-ray of $^{241}$Am in Fig 6. Nevertheless, when the spectrum in Fig. 6 was unfolded analytically, estimates were obtained of $^{239}$Pu and $^{241}$Am lung contents that were not significantly different from the true values. The prominence of the 59.5 keV $\gamma$-ray peak of $^{241}$Am in the Be windows is negligible. It can be seen from these data that a $^{239}$Pu lung content of 0.65 kBq (17.6 nCi) can be measured in one hour with a precision corresponding to twice the square root of the background from an unexposed subject. Our results suggest that if the number of detectors were increased to cover both lungs optimally, the $^{239}$Pu lung content that could be measured in one hour with the same statistical precision would be reduced by less than a factor of two, i.e., $0.4 \times 10^{-1}$ kBq ($10$ nCi).

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Fig. 7 Present orientation of Si(Li) detector array vis-a-vis lung. Other orientations are discussed in text.

reflects the higher sensitivity of the Si(Li) detector array for $^{239}$Pu than for $^{241}$Am. An $^{239}$Pu lung content of 5 Bq (0.14 nCi) can be detected in one hour with a precision corresponding to twice the square root of the background from an unexposed subject. This detectable level is 7 times lower than the $^{241}$Am level shown in Fig. 6 and therefore measurement of $^{241}$Am is expedient for initial screening.

The present configuration of the Si(Li) detector array and its orientation to the subject for lung counting of actinides is shown in Fig. 7.

### Table IV

<table>
<thead>
<tr>
<th>Detector Array Configuration</th>
<th>$^{239}$Pu Relative Count-Rate (Signal)</th>
<th>Unexposed Subject Relative Count-Rate (Bkgd)</th>
<th>Relative Figure of Merit (Signal/√Bkgd)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A. Present Array over R. Lung: parallel to chest</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>B. Present Array across Chest:</td>
<td>0.83</td>
<td>1</td>
<td>0.83</td>
</tr>
<tr>
<td>2 det. over R. lung</td>
<td>2 det. over sternum</td>
<td>2 det. over L. lung</td>
<td></td>
</tr>
<tr>
<td>C. Conceptual Reconfigured Array:</td>
<td>0.87</td>
<td>1</td>
<td>0.87</td>
</tr>
<tr>
<td>4 det. over R. lung</td>
<td>2 det. over L. lung</td>
<td></td>
<td></td>
</tr>
<tr>
<td>D. Conceptual 8-Detector Array:</td>
<td>1.10</td>
<td>1.33</td>
<td>0.95</td>
</tr>
<tr>
<td>4 det. over R. lung</td>
<td>4 det. over L. lung</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
The response of the array for a given counting time in the orientation shown in Fig. 7 is compared in Table IV with the responses for other orientations and conceptual reconfigurations of the array. These other responses were determined by making appropriate measurements with the presently configured array over the phantom thorax loaded with 239Pu lungs. The data of Table IV indicate that for uniformly labeled lungs, precise positioning of the array is probably not critical. Furthermore, these data indicate that there is no signal-to-noise advantage to be gained by increasing the size of the array to 8 detectors.

V. CONCLUSION

A new detector system has been developed that can measure unambiguously in vivo low levels of Pu in the presence of 241Am. The sensitivity of the detector approaches the maximum achievable in a direct measurement. It is shown that the minimum detectable level is primarily limited by the natural radioactivity in the human body. A lung content of 0.65 kBq (18 nCi) 239Pu in a person of average build can be measured reliably in 60 min. Our data indicate that there is no significant advantage to be gained in sensitivity by increasing the array size from 6 detectors over one lung to 8 detectors, where 4 are placed on each lung. It may, however, be desirable to reduce somewhat the size of the NaI(Tl) shield so when the Si(Li) detector array is positioned over the lung as shown in Fig. 7, it does not interfere with the chin. A small reduction in size is not expected to affect the background suppression appreciably. Replacing the NaI(Tl) with a smaller BGO scintillator is an alternative that merits consideration. The contamination in the Be windows can readily be eliminated by replacing the Be with composite windows consisting of multi-layer aluminized Mylar, Kapton, or other plastic film[7,8].

The Si(Li)-NaI(Tl) detector was shown to have a lower background in the UL x-ray energy region than any other detector type currently in use. The Si(Li) background is less than 1/4 of that of a comparable area and resolution planar HPGe detector and 1/15 of that of an equal area NaI(Tl)-CsI(Tl) phoswich system. The salient features of the Si(Li) detector configuration are:

- High energy-resolution in the 15-20 keV x-ray energy range.
High L x-ray detection efficiency equal to the efficiency per unit area of high purity Ge (HPGe) detectors and NaI(Tl)-CsI(Tl) phoswich systems.

- Low intrinsic sensitivity to γ-ray background.
- Anticoincidence background suppression similar to that of phoswich systems.

A complementary report is now being prepared on the application of the new detector system to the practice of health physics. It will present lung-count calibration factors for L x-rays of different energies per unit activity, and will include observed relationships between spectral peak ratios and chest-wall thickness. Estimation of chest-wall thickness is indispensable for accurate determination of actinide lung contents by L x-ray spectrometry.

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References


