A TRIAL PRODUCTION OF PLUTONIUM CONTAMINATED WOUND COUNTERS

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Abstract—A rapid increase of the amount of plutonium to be handled in our Institute has accelerated the need for preparation of wound counters capable of detecting at least a fraction of MPBB (40 nCi) in a plutonium contaminated wound.

Two kinds of wound counters, a xenon gas-filled proportional counter and a cesium iodide crystal scintillation counter, were constructed and their characteristics were examined. The minimum detectable amount for the former was 5.5 nCi and that for the latter was 0.83 nCi when the plutonium was imbedded 6 mm under tissue and the window of the detector was positioned at 3 mm above the skin surface. In order to determine the location of plutonium, a 0.1-mm-thick lead plate and a lead-impregnated flexible material were prepared.

The method of determination of the effective depth of contaminants is discussed below mainly for the cases of pure plutonium and plutonium with an unknown content of americium in the wound. Furthermore, we have shown that the depth distribution of the contaminants can be obtained in fairly good agreement with the actual distribution by the method of expansion of source distribution using the orthonormal function series method. This method will be valid provided the contaminant lies continuously in the wound.

1. INTRODUCTION

Following the rapid increase of the amount of plutonium handled in the plutonium laboratory in our Institute, the preparation of wound counters has become an urgent necessity in order to determine the location, the depth and the amount of plutonium in wounds when such an accident occurs and surgical excision of skin and tissue is required. It is necessary to detect at least a fraction of the maximum permissible body burden (MPBB = 40 nCi) in a plutonium contaminated wound, without cutting or probing, in order to aid the decontamination operation and the medical treatment of personnel who sustain such injuries. Similar counters, especially the scintillation types, were already developed in several other countries.[1-4] But, considering the cases of pure plutonium or unknown content of americium in plutonium to be handled, a xenon gas-filled proportional counter was examined to determine the effective depth of contaminants in wounds using the photopeak ratio of L_α–X and L_β–X rays emitted from plutonium. A cesium iodide crystal scintillation counter was also examined to determine the effective depth using L–X and γ-radiation. Other characteristics of these two counters were compared.

In order to determine the location of contaminants, a 0.1-mm-thick lead plate or a lead-impregnated flexible material were prepared to perform grid surveys. A transparent material is preferable.

Furthermore, the development of a code for computing the depth distribution of contaminants in a wound was carried out by means of an expansion of the source distribution using the orthonormal function series method.[5]

2. EXPERIMENTAL ARRANGEMENTS

A proportional type detector, Aloka PC-101 Xe tube filled with xenon gas containing 10 per cent methane gas under a pressure of 32 cm Hg and having a 1.8 mg/cm^2 mica window, was used. The mica window was covered by an aluminium foil with sufficient thickness to

cut off α-rays emitted by plutonium. The sensitive volume of the counter is 24 mm (diameter) $\times$ 100 mm.

The scintillation type detector probe assembly contains a cesium iodide scintillation crystal, 1 in. in diameter and 1 mm in its thickness, with an aluminium foil having sufficient thickness to cut off α-rays. The crystal was mounted on a Toshiba 7696 photo-multiplier tube.

These detectors were used together with a TMC 400 channel pulse height analyzer.

In both cases L–X rays produced by the internal conversion of 37- and 51-keV gamma radiation associated with the alpha decay of plutonium-239, the energy ranging from 13.6- to 20.5-keV and having an emission rate of 3.8% against alpha decay, were used because the 51-keV gamma-ray was difficult to detect.

Acryl acid resin plates having various thicknesses and with specific gravity in the range 1.16–1.2 were used as tissue equivalent material instead of human tissue to measure the absorption curves of the X-rays and thus assess an effective depth of plutonium in tissue.

The standard source of plutonium-239 made in the Radiochemical Centre (RCC) in England having activity equal to $(8.897 \pm 0.01) \times 10^{-2}$ μCi and distributed on a platinum plate for about 3 mm in diameter, and a standard source of americium-241 made in the radiochemical laboratory in JAERI having the activity of 0.5 μCi in alpha decay were used.

3. RESULTS AND DISCUSSIONS

3.1. *Proportional Counter Method*

A plateau curve and a background counting rate of the xenon gas-filled proportional counter were measured using an americium-241 standard source in order to obtain an adequate working high voltage with background counting as low as possible, as shown in Fig. 1. From Fig. 1, the working high voltage of 1600 V was selected, the background counting rate for the whole energy range being about 54 cpm.

Figure 2 shows the relationship between the channel number of the multichannel pulse height analyzer and the X-ray energy when the xenon gas-filled proportional counter was operated under the high voltage of 1600 V. Standard sources of barium-133 and selenium-75 were used to plot the curve.

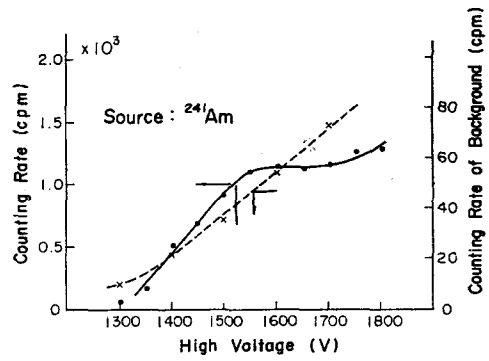

FIG. 1. Plateau and background curves of Xe gas-filled proportional counter.

Figure 3 shows the X-ray spectrum of plutonium-239 obtained by the proportional counter. The second and the third photopeaks are considered as L_α– and L_β–X rays emitted by the plutonium-239 source, respectively, on the basis of the channel number to the pulse height–energy dependence curve in Fig. 2. The distance between the source and the window of the proportional counter was 3 mm. The first peak lying at the energy of about 6 keV was investigated by comparing the spectra with and without absorber as shown in Fig. 4. The lead plate having 0.1 mm thickness was used as an absorber and inserted between the americium-241 source and the window of the counter. It is obvious from the figure that the occurrence of the first peak is not due to emission from the source because, in spite of the disappearance of the peaks of the L_α– and

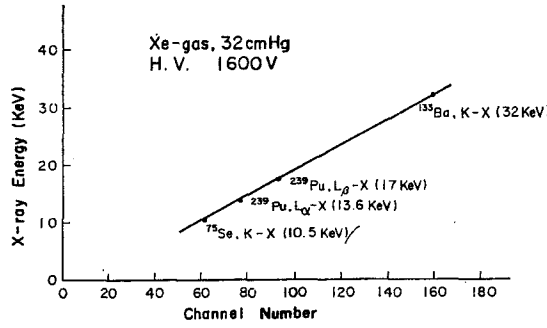

FIG. 2. Relation between pulse height and energy of Xe gas-filled proportional counter.

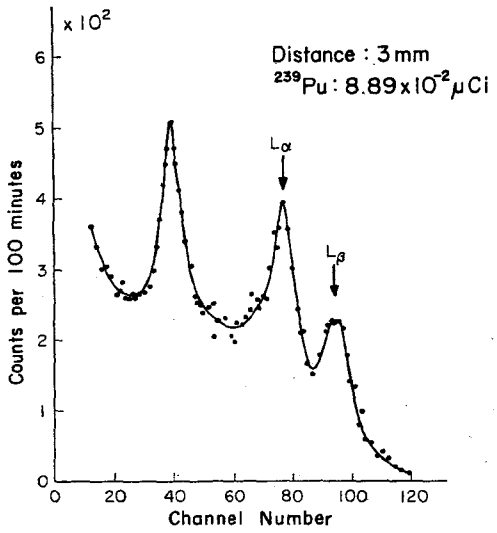

FIG. 3. X-ray spectrum of ^{239}Pu measured by Xe gas-filled proportional counter.

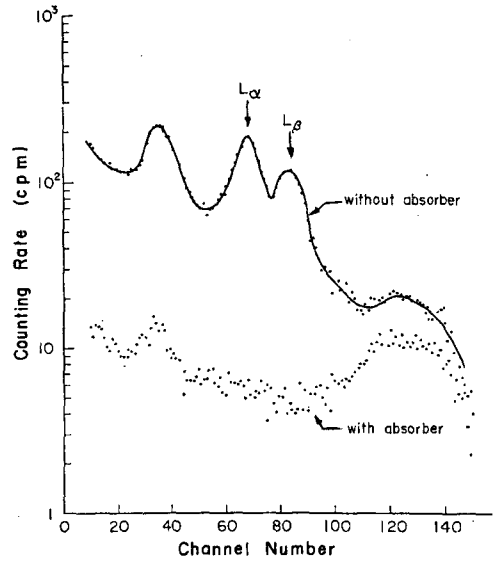

FIG. 4. X-ray spectra of ^{241}Am with and without absorber of 0.1 mm thick Pb.

L_β–X rays with absorber present, the first peak with lower energy remained almost unchanged.

The attenuation coefficients of L_α– and L_β–X rays in tissue differ according to their energy as shown in Fig. 5, for example. In this case, a 3.1 mm thick acryl acid resin plate was inserted between the americium-241 source and the window of the counter. Curves (a) and (b) in Fig. 5 show the spectra of the americium-241 source without and with the 3.1 mm thick acryl acid resin plate, respectively.

It is easy to calculate the depth of contaminants in tissue in the case of a point source by using results obtained for the arrangements shown in Fig. 6 (a) and (b). Fig. 6 (a) shows the case when the contaminants were imbedded at the depth x under the skin surface and the end of detector was positioned at the distance t from the skin surface. The counting rates and the attenuation coefficients of X-rays are designated by N_α and μ_α for the L_α–X ray and by N_β and μ_β for the L_β–X ray, respectively. If the tissue in Fig. 6 (a) was removed, the counting rates will be designated $N_{\alpha o}$ for the L_α–X ray and by $N_{\beta o}$ for the L_β–X ray, and the attenuation coefficient for air is nearly equal to unity, as indicated in Fig. 6 (b). In both cases the geometry remains the same. The following relationships hold with these notations:

$$\left.\begin{aligned} N_\alpha &= N_{\alpha o} e^{-(\mu_\alpha)x} \\ N_\beta &= N_{\beta o} e^{-(\mu_\beta)x} \end{aligned}\right\} \qquad (1)$$

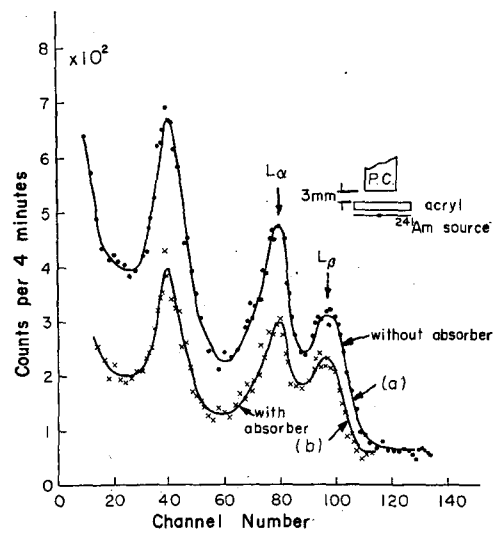

FIG. 5. X-ray spectra of ^{241}Am without and with absorber of acryl (3.1 mm).

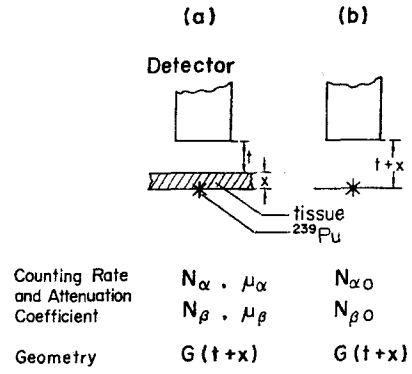

FIG. 6. Schematic arrangement of detector and source.

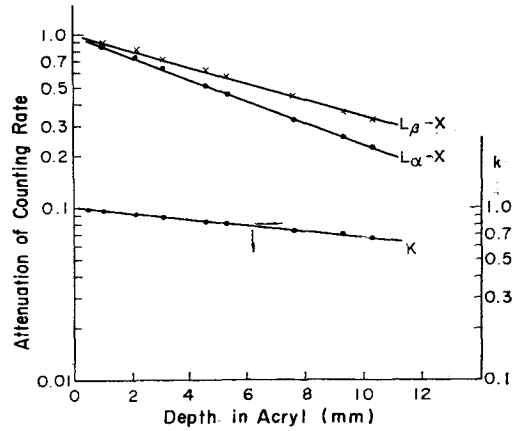

FIG. 7. X-ray attenuation curves in acryl measured by Xe gas-filled proportional counter.

If we put,

$$K = (N_\alpha/N_\beta)/(N_{\alpha o}/N_{\beta o}) = (N_\alpha/N_{\alpha o})/(N_\beta/N_{\beta o}) \quad (2)$$

then,

$$\ln K = -(\mu_\alpha - \mu_\beta)\, x. \quad (3)$$

If we consider that the values μ_α and μ_β for the tissue are constant, the depth x can be calculated from equation (3) when the value K is known. In Fig. 7, the straight lines L_α–X and L_β–X show the values $N_\alpha/N_{\alpha o}$ and $N_\beta/N_{\beta o}$ of equation (2), respectively, when the thickness of the acryl acid resin plates is changed, and the straight line K shows the change of the ratio $(N_\alpha/N_{\alpha o})/(N_\beta/N_{\beta o})$ with the increase of the thickness of the plate. In practice, N_α and N_β can be obtained easily from measured values for the contaminated wound with the proportional counter, and the value K by dividing the ratio N_α/N_β by the value $N_{\alpha o}/N_{\beta o}$ measured beforehand. Then the depth of the point source contaminant x can be obtained from the straight line K in Fig. 7, and the amount of contaminant in the wound can also be obtained from the peak value of the spectrum. At the same time the location of the contaminant can easily be determined by a grid survey method using a 0.1-mm-thick lead plate or a lead-impregnated material.

But, it is not practical to consider that the contaminants always lie in the wound in such a single point source. It is rather usual that the contaminants are distributed in several points in the wound in the case of insoluble contaminants, and distributed continuously in the case of soluble contaminants. In such cases, the estimated value of the depth x will become an effective depth of the whole contaminants. It is desirable to know the exact depth distribution of contaminants quickly, if possible, in order to perform an effective surgical excision of skin or tissue. Consequently, the development of a code for computing the depth distribution of contaminants in a wound was carried out theoretically.

Let us consider that contaminants having unit activity are imbedded in tissue at various depths x and the spectra having n peaks were measured from outside the skin surface. Furthermore, let us put $g_i(x)$ for the counting rates of the ith peak for various x's. Then, it is possible to obtain an orthonormal function set $\{\phi_i(x)\}$ from the response group $\{g_i(x)\}$ by applying the Gram-Schmidt process. The relationship can be represented by a matrix G as follows:

$$G = B \cdot \psi \quad (4)$$

where G is an n-column matrix having $\{g_i(x)\}$ as the elements, B is a conversion matrix $(n \times n)$, and ψ is also an n-column matrix having $\{\phi_i(x)\}$ as the elements.

Now, let us represent the distribution function of the contaminants along the depth x of the wound by $s(x)$. Assuming that the function $s(x)$ can be expanded by the orthonormal function set $\{\phi_i(x)\}$, we have again the following matrix equation;

$$s(x) \cong A \cdot \psi \tag{5}$$

where A is an unknown conversion matrix. If we denote the measured value of the ith peak of the spectrum by n_i, it follows that

$$n_i = \int g_i(x) \cdot s(x)dx.$$

Therefore,

$$N \cong B \cdot \int \psi \cdot \psi^T dx \cdot A^T$$
$$= B \cdot W \cdot A^T \tag{6}$$

where N is a column matrix having $\{n_i\}$ as the elements, and W is $\int \psi \cdot \psi^T dx$. From equation (6), we have

$$A \cong (W^{-1} \cdot B \cdot N)^T. \tag{7}$$

Substituting from equations (4) and (7) into equation (5), we have

$$s(x) \cong N^T \cdot (B^{-1})^T \cdot (W^{-1})^T \cdot B^{-1} \cdot G. \tag{8}$$

It is obvious from equation (8) that the approximate distribution of contaminants can be obtained by the combination of the response function G weighted by the counting rate N.

In order to confirm the theoretical results experimentally, a 0.5 μCi americium source was used to obtain the spectra for americium imbedded at various depths under tissue. The $g_i(x)$'s for the counting rates of L_α–, L_β– and L_γ–X rays of the spectra were plotted as shown in Fig. 8. Using these $g_i(x)$'s, two different kinds of depth distributions $s(x)$'s which were assumed as the solid lines in Fig. 9, were calculated by IBM computer. The calculated $s(x)$'s are shown by the dotted lines in the same figure. It is obvious from the figure that the calculated distributions agree fairly well with the assumed

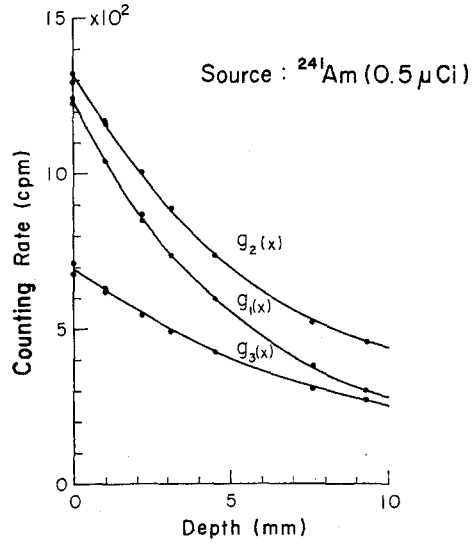

FIG. 8. Depth dependence curves of counting rate for L_α–, L_β– and L_γ–X rays.

distributions of contaminant in the case of a continuous distribution, but presumably it is difficult to obtain such a successful agreement in the case of discontinuous distribution.

3.2. *Scintillation Counter Method*

Another experiment using a thin cesium iodide scintillation crystal was carried out in order to compare with the results obtained

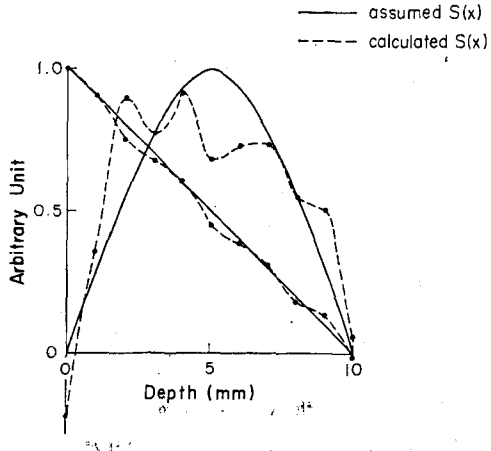

FIG. 9. Assumed and calculated depth distributions of contaminants (^{241}Am).

with the xenon gas-filled proportional counter. Figure 10 shows the X- and γ-rays spectra of americium-241 and plutonium-239 obtained by the cesium iodide scintillation crystal, 1 in. in diameter and 1 mm thick. It is obvious from the figure that the plutonium-239 source has a small content of americium-241.

Here again we have examined the sensitivity for determining the depth of contaminants using

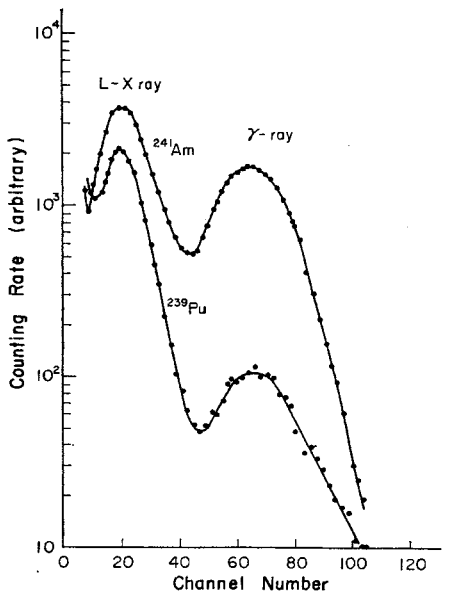

FIG. 10. X- and γ-ray spectra of ^{241}Am and ^{239}Pu measured by CsI(T1) crystal.

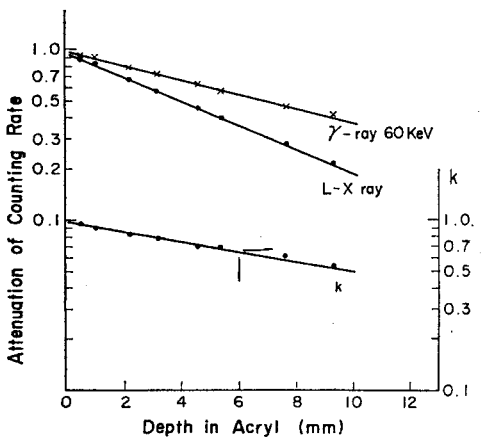

FIG. 11. X- and γ-ray attenuation curves in acryl measured by CsI (T1) crystal.

L–X and γ-rays (60 keV) emitted from the americium-241 source and acryl acid resin plates having various thicknesses for absorber, as shown in Fig. 11. By comparing Figs. 7 and 11, it is clear that the sensitivity for determining the depth of contaminants is greater for the scintillation counter than for the proportional counter owing to the utilization of the higher energy 60 keV γ-radiation. The minimum detectable amounts for the proportional counter and for the scintillation counter were 2.5×10^{-3} and 3×10^{-4} μCi, respectively, when the contaminants were on the skin surface and 5.5×10^{-3} and 8.3×10^{-4} μCi, respectively, when the contaminants were at the depth of 6 mm in the tissue equivalent acryl acid resin plate, for a measuring time of 10 min with a confidence limit of 95% if the window of the detector were positioned at 3 mm above the skin surface, as shown in Table 1. From these results, a scintillation counter appears to be more effective than a proportional counter, but the proportional counter is useful when the orthonormal function series method is used to obtain the depth distribution of contaminants in a wound, especially in the case of a contaminant composed of pure plutonium.

4. CONCLUSION

Because of the urgent need for wound counters in order to determine the location, the depth and the amount of plutonium in wounds as an aid in determining the need for surgical excision of skin and tissue, a xenon gas-filled proportional counter was examined as a means of making effective depth estimation when the wounds were contaminated by pure plutonium. A thin cesium iodide crystal type scintillation counter was also examined to compare its sensitivity for determining the effective depth and the minimum detectable amount of contaminants in a wound with the comparable results using the proportional counter when the wounds were contaminated both by plutonium and americium.

In the case of a contaminant including both plutonium and americium, the sensitivity for effective depth determination and the minimum detectable amount were better for the scintillation counter than for the proportional coun-

Table 1. *Characteristics of detectors.*
(^{239}Pu source: 8.89×10^{-2} μCi)

Detector	Proportional counter	CsI (Tl) crystal 1 in. dia × 1 mm thick
Distance to source (mm)	3	3
Energy range of L-X ray (keV)	10.5–22	10.5–22
Net counting rate (cpm)	106	2482
Background (cpm)	9	67
M.D.A.* (μCi)		
Skin surface	2.5×10^{-3}	3×10^{-4}
Under tissue (6 mm)	5.5×10^{-3}	8.3×10^{-4}

* Measuring time, 10 min.
Limit of confidence, 95%.

ter, but the effective depth estimation was only possible by use of the proportional counter when the contaminant includes only pure plutonium.

Furthermore, the development of a code for computing the depth distribution of contaminants in a wound was carried out, theoretically, by means of an expansion of results obtained by the counters, using the orthonormal function series method. For this purpose, the high resolution of the proportional counter is needed to make it successful. A verification of this method was performed using measured $\{g_i(x)\}$ values, and has shown that the two calculated distribution curves of the contaminants agree fairly well with the assumed distribution curves.

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