THE DETECTION AND MEASUREMENT OF PLUTONIUM CONTAMINATION IN WOUNDS

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Abstract—Plutonium is extensively used in atomic research and industry and inevitably there occur wounds and injuries which contain plutonium. Because of its high toxicity it is important to identify, locate and measure any plutonium present. At levels which have medical significance, the quantities involved are minute, but there is sufficient emission of accompanying X- and gamma radiations to permit identification and assay by radiometric techniques.

X-rays in the band 13 to 20 keV, gamma rays of about 60 keV and gamma rays of about 384 keV can all be used for this work: the problems and the limitations associated with measurements in these three energy bands are discussed in the paper, including the usefulness of energy spectrometry to eliminate errors due to self absorption.

There are various instruments available, and usable, for radiometri determinations of plutonium in wounds. Several of these are described with their limitations and detection capabilities.

A case of a contaminated wound is quoted to illustrate some of the techniques and problems in the assessment of plutonium content. Some subsequent work undertaken to improve identification and measurement is also described.

A number of specific actions are suggested, which should be taken, following the occurrence of a wound to ensure that any plutonium present before and after medical treatment is correctly identified and measured.

1. INTRODUCTION

In working with plutonium and its compounds, one route by which radioactive material is accidentally introduced into the body is through minor wounds. These are usually small cuts, punctures and splinters in the hands, caused by objects sharp enough to penetrate through box gloves and through the skin. A few cases have been reported of more serious wounds resulting from accidents in plutonium workplaces. At the AWRE, persons working with radioactive materials are instructed to report all minor injuries which occur in controlled areas. Over a period of 18 months 225 injuries were reported. In 23 cases plutonium contamination was suspected and in 4 of these cases plutonium contamination was confirmed.

The available evidence appears to show that a small fraction of the plutonium injected as metal in minor injuries finds its way into the circulating body fluid within the first few days but most of the material remains at the site of the wound for a long time unless excised or sloughed off with scab.[1] It is generally considered desirable to remove as much as possible of the plutonium from superficial wounds in order to reduce the risk of dispersion in the body and to prevent high dose to local tissue. The maximum permissible body burden of ^{239}Pu when the bone is considered to be the critical organ is only 0·04 μCi (0·64 μg). A single piece of plutonium of this mass has a diameter of only about 40 μm.

There is thus a need to apply suitable radiation measuring techniques to:

(a) confirm and assess the initial contamination,
(b) locate the site of contamination accurately,
(c) demonstrate the success of surgical operations, and
(d) study the fate of any plutonium remaining at the wound site after surgical treatment.

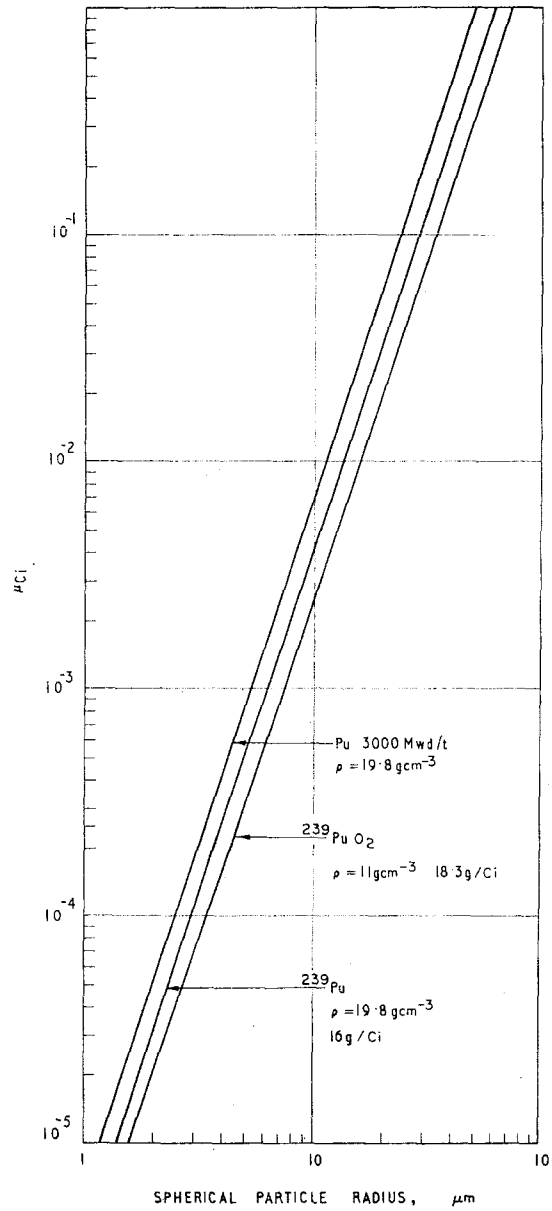

FIG. 1. The radioactivity of spherical particles.

2. MONITORING PRINCIPLES

Personal monitoring for plutonium contamination is usually carried out with a standard alpha particle detector but, owing to the short range of the alpha particles in tissue (about 40 μm), this method will not detect plutonium which has penetrated the outer skin layer or which is covered over with blood. If an alpha probe alone is used to check minor wounds after they have been washed, there is a distinct risk that persons will resume work while still carrying significant contamination below the skin. It has therefore become accepted practice to measure the uranium L X-ray emission at about 17 keV, which follows alpha decay in a small percentage of the disintegrations. Table 1 shows the principal energies and abundance of these emissions for disintegrations of plutonium isotopes and daughter products. The detectors employed are scintillation counters containing thin flat sodium iodide (Tl) crystals covered by aluminium or beryllium foils. These are discussed in more detail below.

Interpretation of the measurements at 17 keV may be complicated by absorption of the X-rays in overlying tissue, by self-absorption in particles of the contaminant, and by the presence of radio-nuclides other than ^{239}Pu which also emit both alpha-particles and X-rays. Tyler[2] has examined the problem of self-absorption for ^{239}Pu in the form of metal and certain compounds. He has shown that, because self-absorption of alpha particles is generally greater than self-absorption of 17 keV X-rays, monitoring with an X-ray probe does not underestimate the potential dose to tissue at the site of the wound but may lead to an underestimate of the amount of plutonium available for transfer to other parts of the body.

In addition to the uranium X-rays there are other penetrating radiations which may be emitted in sufficient quantity to yield useful information; notably the 60 keV gamma-rays of ^{241}Am (daughter of ^{241}Pu) and the 384 keV gamma-rays of ^{239}Pu. The latter are of very low abundance (0·0012%) but in the case of a very severely contaminated wound, could be used to determine an upper limit for the ^{239}Pu content, rapidly and with the minimum uncertainty of interpretation. The practical usefulness of measurements at the different energies is further discussed below.

3. THE TYPES OF MEASUREMENT REQUIRED

3.1. *Initial Monitoring*

Initial monitoring of suspected punctures and other very minor wounds is likely to be carried out, after washing, at the place of work or at the nearest health physics office, using a portable

Table 1. *Principal X- and γ-rays accompanying Disintegrations of Plutonium Isotopes.*

Isotope	Half-life (days)	α particles per sec per g of Pu isotope	No. of photons emitted per alpha particle emission					
			12–14 keV	17–18 keV	20–21 keV	52–65 keV	100 keV	384 keV
^{238}Pu	$3{\cdot}3 \times 10^{4}$	6×10^{11}		$13{\cdot}5 \times 10^{-2}$			$8{\cdot}4 \times 10^{-5}$	
^{239}Pu	$8{\cdot}9 \times 10^{6}$	$2{\cdot}25 \times 10^{9}$	$1{\cdot}3 \times 10^{-2}$	$1{\cdot}9 \times 10^{-2}$	$3{\cdot}15 \times 10^{-3}$	7×10^{-5}	$5{\cdot}2 \times 10^{-5}$	$1{\cdot}2 \times 10^{-5}$
^{240}Pu	$2{\cdot}4 \times 10^{6}$	$8{\cdot}3 \times 10^{9}$		11×10^{-2}				
^{241}Pu	$4{\cdot}8 \times 10$	$9{\cdot}6 \times 10^{7}$					·35	
^{237}U*	6·75		$3{\cdot}55 \times 10^{-2}$	$8{\cdot}15 \times 10^{-2}$	$2{\cdot}6 \times 10^{-2}$	·385	·56	
^{241}Am†	$1{\cdot}7 \times 10^{5}$	$5{\cdot}9 \times 10^{9}$	$14{\cdot}8 \times 10^{-2}$	$19{\cdot}7 \times 10^{-2}$	4×10^{-2}	·38	·315	

* The figures relate to the amount of ^{237}U in equilibrium with 1 g of ^{241}Pu and represent the number of photons per ^{241}Pu alpha emission.
† The figures relate to the amount of ^{241}Am present in 1 g of ^{241}Pu 1 year after separation.

or semi-portable instrument. It is important that contamination under the skin should not be missed at this stage so the monitoring must be performed carefully and slowly, since the count-rates for significant contamination are quite low and the X-ray probes have a sensitive area of only a few square centimetres. Certainty of detection is more important than precision of assessment at this stage and the lower limit of detection for ^{239}Pu ideally should be 0·005 μCi or less. It would assist in ensuring a really adequate check, if the whole hand could be monitored at once in an X-ray monitor for a fixed time interval in much the same way as with a normal α/β hand contamination monitor. No simple version of this appears to have been produced. A well-type phosphor large enough to monitor a whole finger has been tried, but so far, it has not been possible to make an inexpensive detector of this form sufficiently robust and sensitive for general use. Furthermore it would not cope with the small proportion of wounds which occur to the palm of the hand.

3.2. *Accurate Location of Contamination*

When a contaminated wound has been confirmed, it will be referred to the medical officer for treatment. A more precise measurement of the amount and location of the plutonium is now necessary. Accurate location by X-ray measurements is quite difficult because the count-rate per microcurie is very low and any degree of collimation of the detector reduces it still further. It has been found best to mount the detector in a stand about 5 mm above the wound at a point which gives the maximum count-rate and then slide straight-edged or iris-shaped absorbers over the skin, noting the points at which the count rate is sharply diminished.

3.3. *Precise Determination*

In the case of very superficial wounds there is generally no problem about excising the foreign matter. In a more serious case the medical officer may have to balance the desirability of removing all the material, against the possible damage to the individual as a result of surgical operation. In such a case he will require a fairly accurate estimation of the amount of plutonium involved. To achieve this, it is desirable to measure the spectrum of radiation emitted from the wound, and, if possible, obtain a sample of the relevant plutonium from which to prepare a reference source. The reference source should be a thinly deposited source suitable for alpha counting and alpha spectrometry in addition to gamma spectrometry. Corrections can then be made for the presence of isotopes other than ^{239}Pu and for self-absorption.

Any material which is excised from the wound is checked for activity using the X-ray detector. It may then be desirable to retain this material as an aid to further studies.

Finally when the wound has been treated, an attempt should be made to determine an upper limit for the residual contamination. Here again a knowledge of the radiation spectrum can assist towards obtaining a more accurate estimate.

4. INSTRUMENTS AVAILABLE FOR WOUND MONITORING

Several instruments suitable for the detection of X-rays from plutonium have been described.[3-5] The following have been used at the AWRE.

4.1. *Portable Contamination Monitor Type 1320X*

This employs a 55 mm diam. scintillation probe fitted with a sodium iodide phosphor 36 mm diam. × 1·0 mm thick under a 0·05 mm aluminium foil. The probe is coupled to a battery-powered portable ratemeter which, when used in the X-ray monitoring role, has an energy "window" extending from 11 keV to 23 keV approximately. The instrument is designed for general contamination survey duties in the field and has a background count-rate of about 20 counts/min when unshielded. It is used in laboratories for initial checks on suspected skin punctures. Under these conditions, the lower limit of detection is about 0·01 μCi ^{239}Pu. A similar probe gave a background of about 1 count/min in the AWRE low-background facility.[6]

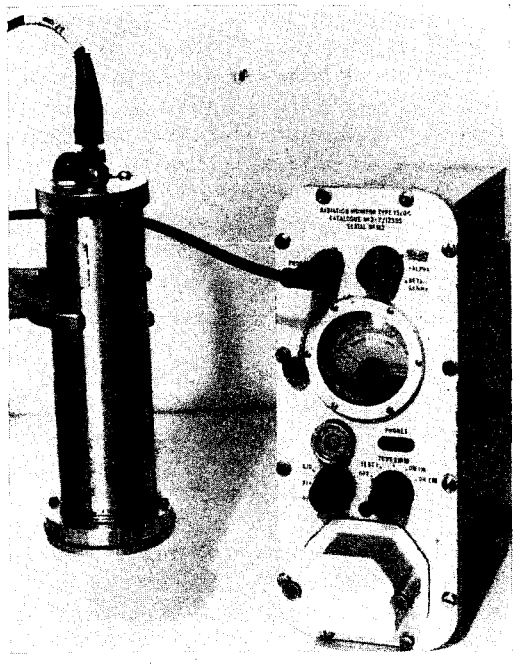

FIG. 2. Portable contamination monitor Type 1320 X.

4.2. *Plutonium Wound Monitor Type PNI 1080**

This instrument employs a 35 mm diam. probe fitted with a sodium iodide phosphor 10 mm diam. × 1·0 mm thick covered with a 0·05 mm aluminium foil. The small, mains-powered ratemeter incorporates an electro-mechanical register for use at low pulse-rates. The circuit has an energy "window" extending from 13 keV to 21 keV approximately. In normal laboratory environments the instrument has a background count-rate of about one count per minute and the lower limit of detection is about 0·005 μCi ^{239}Pu. It is used in our medical and health physics centres for measurement and accurate location of plutonium in wounds.

4.3. *Selective Gamma Monitor Type NIS 322 (Pu)*

In this instrument the scintillation detector, ratemeter and indicator are incorporated in a single pistol-grip unit which can be held comfortably in one hand. In its normal form it employs a phosphor 25 mm thick × 37 mm diam. and is "gated" to accept the 384 keV gamma-rays from ^{239}Pu. The lower limit of detection, in a normal environment is about 500 μCi (about 8 mg) at 5 cm from the end of the detector. Its role in wound monitoring

* Manufactured by Plessey Nucleonics Ltd., Northampton, England.

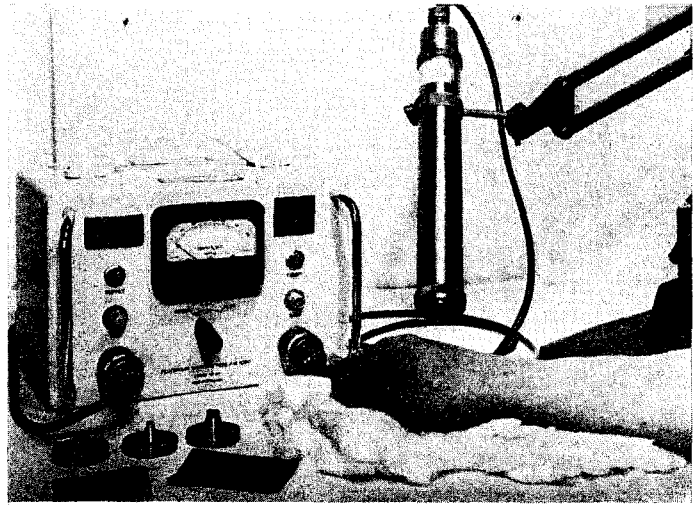

FIG. 3. Plutonium wound monitor Type PNI 1080.

would therefore be confined to a very serious accident where the amount of plutonium involved might imply considerable self-absorption of the low energy X-rays.

Better detection of the 384 keV radiation can be achieved with improved geometry. A 75 mm diam. crystal, having a central well and mounted in a 50 mm lead shield, enabled measurements to be made on a dummy finger, down to 2 μCi ^{239}Pu in a normal laboratory.

Because of the convenient shape and reliable operation of the Selective Gamma Monitor Type NIS 322, an experimental version of it has been constructed for measurements at low energy (Fig. 4). The dimensions of the scintillator and aluminium foil are the same as those of the Probe Type 1320X. The electronic circuit incorporates an energy window which can be switched to either of the ranges:

12 keV–22 keV (for uranium *L* X-rays)
or 48 keV–72 keV (for 60 keV emission from ^{241}Am).

It is hoped that this instrument will replace the Monitor Type 1320X for plutonium X-ray surveys.

4.4. *Use of Multi-channel Analysers*

If one of the thin-crystal scintillation probes is connected to a multi-channel analyser, the energy spectrum from about 12 keV to about 100 keV can be plotted for the radiation from the wound. The presence of ^{241}Am is then revealed by the peak at 60 keV. It may take about 20 min to obtain a useful spectrum so the subject must be positioned comfortably. It is useful to mount the scintillation probe in a simple lead castle into which the hand can be inserted conveniently or to carry out the measurement within a low background facility such as a whole-body counting room.

5. THE EFFECT OF ISOTOPES OTHER THAN ^{239}Pu

If the plutonium has undergone considerable reactor irradiation it will contain, in addition to ^{239}Pu, proportions of the alpha-emitters ^{238}Pu and ^{240}Pu and the beta emitter ^{241}Pu (half-life 13 years). After separation of pure plutonium from irradiated fuel, ^{237}U and ^{241}Am, the daughters of ^{241}Pu, will begin to grow. The short-lived ^{237}U (6·75 days) reaches equilibrium within a few weeks. ^{241}Am (458 years) will increase almost linearly with time for a few years after separation.

The presence of isotopes other than ^{239}Pu increases the alpha emission per gramme of material; however it increases even more the emission of low energy X-rays per gramme of material, so the significance of the X-ray measurements as an indicator of internal radiation hazard changes as high irradiation and long americium growth are attained. Table 2 shows that for plutonium which has undergone irradiation of 3,000 MWd/t followed by one year's

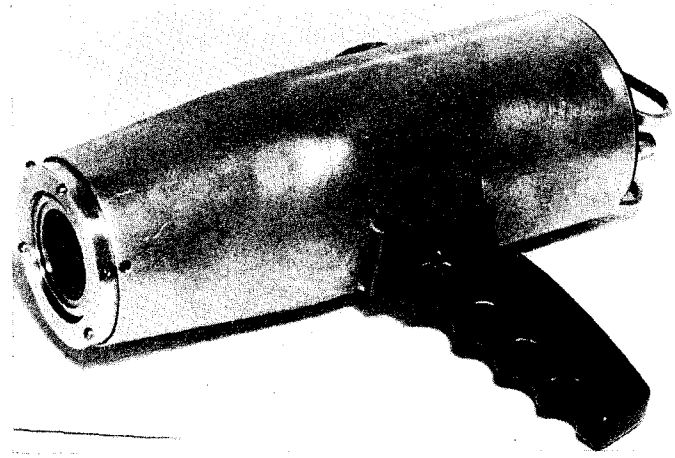

FIG. 4. Prototype of selective gamma monitor Type NIS 322 modified for X-ray monitoring.

Table 2. Emissions from 1 g of Plutonium 1 Year after Separation.

Irradiation / Isotope	500 MWd/t				3000 MWd/t				6000 MWd/t			
	Typical abundance w/o	α/s	*X-rays/s	60 keV γ/s	Typical abundance w/o	α/sec	*X-rays/s	60 keV γ/s	Typical abundance w/o	α/s	*X-rays/s	60 keV γ/s
^{238}Pu	0·0052	$3{\cdot}12 \times 10^{7}$	$4{\cdot}2 \times 10^{6}$		0·065	$3{\cdot}9 \times 10^{8}$	$5{\cdot}2 \times 10^{7}$		0·25	$1{\cdot}5 \times 10^{9}$	$2{\cdot}1 \times 10^{8}$	
^{239}Pu	96	$2{\cdot}15 \times 10^{9}$	$6{\cdot}8 \times 10^{7}$		82·6	$1{\cdot}86 \times 10^{9}$	$5{\cdot}85 \times 10^{7}$		69	$1{\cdot}55 \times 10^{9}$	$4{\cdot}9 \times 10^{7}$	
^{240}Pu	3·8	$3{\cdot}15 \times 10^{8}$	$3{\cdot}5 \times 10^{7}$		13·7	$1{\cdot}14 \times 10^{9}$	$1{\cdot}26 \times 10^{8}$		24·5	$2{\cdot}0 \times 10^{9}$	$2{\cdot}25 \times 10^{8}$	
^{241}Pu	0·22	$2{\cdot}1 \times 10^{5}$			3·3	$3{\cdot}15 \times 10^{6}$			5·6	$5{\cdot}3 \times 10^{6}$		
^{237}U			$3{\cdot}0 \times 10^{4}$	$8{\cdot}2 \times 10^{4}$			$4{\cdot}5 \times 10^{5}$	$12{\cdot}2 \times 10^{5}$			$7{\cdot}6 \times 10^{5}$	$2{\cdot}1 \times 10^{6}$
^{241}Am		$1{\cdot}3 \times 10^{7}$	$5{\cdot}2 \times 10^{6}$	$5{\cdot}0 \times 10^{6}$		$1{\cdot}94 \times 10^{8}$	$7{\cdot}7 \times 10^{7}$	$7{\cdot}4 \times 10^{7}$		$3{\cdot}3 \times 10^{8}$	$1{\cdot}32 \times 10^{8}$	$1{\cdot}27 \times 10^{8}$
Totals		$2{\cdot}5 \times 10^{9}$	$1{\cdot}1 \times 10^{8}$	5×10^{6}		$3{\cdot}58 \times 10^{9}$	$3{\cdot}14 \times 10^{8}$	$7{\cdot}5 \times 10^{7}$		$5{\cdot}38 \times 10^{9}$	$6{\cdot}16 \times 10^{8}$	$1{\cdot}3 \times 10^{8}$
Total X-rays/α		$4{\cdot}4 \times 10^{-2}$				$8{\cdot}75 \times 10^{-2}$				$11{\cdot}4 \times 10^{-2}$		
Total 60 keV/α		2×10^{-3}				$2{\cdot}1 \times 10^{-2}$				$2{\cdot}43 \times 10^{-2}$		

*12–21 keV photons.

growth of ^{241}Am, the X-ray measurements may overestimate the alpha hazard by a factor of 2 if the instrument were calibrated with low irradiation plutonium.

The effect of further increases in isotopic content can be determined from the information in Table 1.

6. THE EFFECT OF DEPTH OF PLUTONIUM IN THE WOUND

If the plutonium is injected well below the surface of the skin, the measurements will be affected by absorption of the radiation in the overlying tissue and by the increased distance between source and detector. For minor hand wounds these effects are unlikely to be significant. The X-rays with an average energy of 17 keV have a half-value thickness in tissue of about 7 mm while at 60 keV the half-value thickness is about 35 mm. The geometrical effect is greatest when the detector is placed very close to the skin. Figure 9 shows the effect of separating the source and detector by layers of approximately tissue-like material.

7. EFFECT OF SELF-ABSORPTION IN THE PARTICLES OF CONTAMINANT

Tyler has shown[2] that, if the activity in a wound is concentrated in a single spherical particle, self-absorption of the X-rays may be appreciable for total activities greater than about 10^{-4} μCi ^{239}Pu. When the total activity is about 1 μCi ^{239}Pu, self-absorption may lead to underestimation by an order of magnitude.

Similar calculations have been performed for the 60 keV emissions of ^{241}Am and show (Fig. 5) that self-absorption does not become significant until the single particle size corresponds to about 60 μm diameter (0·1 μCi ^{239}Pu). Hence, if the radiation spectrum from the wound can be plotted so as to include both the X-ray peak and a discernible 60 keV peak, the ratio of these peaks, for a given isotopic composition will indicate the amount of self-absorption. In practice the isotopic composition may not be known, but it may be possible to obtain a sample of the relevant material, from which to prepare a thin source of negligible self absorption. The ratio $\frac{17\text{ keV peak}}{60\text{ keV peak}}$ for the wound spectrum can then be compared with the same ratio for the thin source. Figure 6 shows how the ratio should theoretically be reduced with increasing particle size.

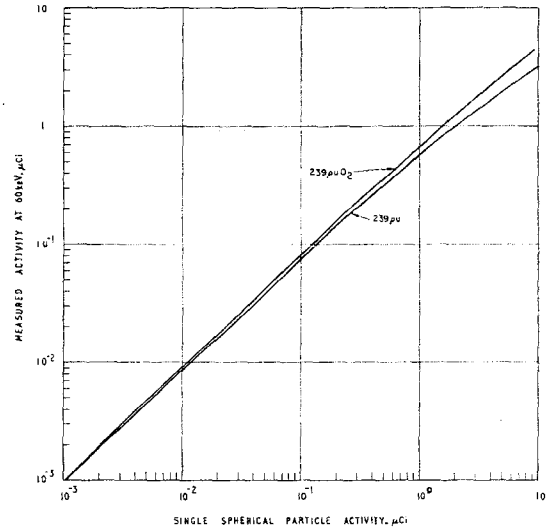

FIG. 5. Correction curves for self-absorption of 60 keV gamma radiation in spherical particles.

8. DESCRIPTION OF A CASE OF A CONTAMINATED WOUND

The following case history of a wound has been selected because it illustrates a number of the points made above.

8.1. *Initial Circumstances*

While manipulating plutonium metal with gloves the subject, "A", received a minor puncture wound close to the finger nail on the side of the middle finger, left hand. By error the hand was monitored only with an alpha probe. This gave a count rate of about 10 counts/s, which was reduced to less than 2 counts/s by washing. "A" then returned to work.

49 days later, on the evidence of urine analysis results, attention was drawn to "A" again and the finger was monitored using X-ray monitors Type PNI 1080 and Type 1320X, both calibrated against a standard plutonium source set in "Perspex". These measurements yielded an estimate of 1.6 μCi of plutonium alpha activity.

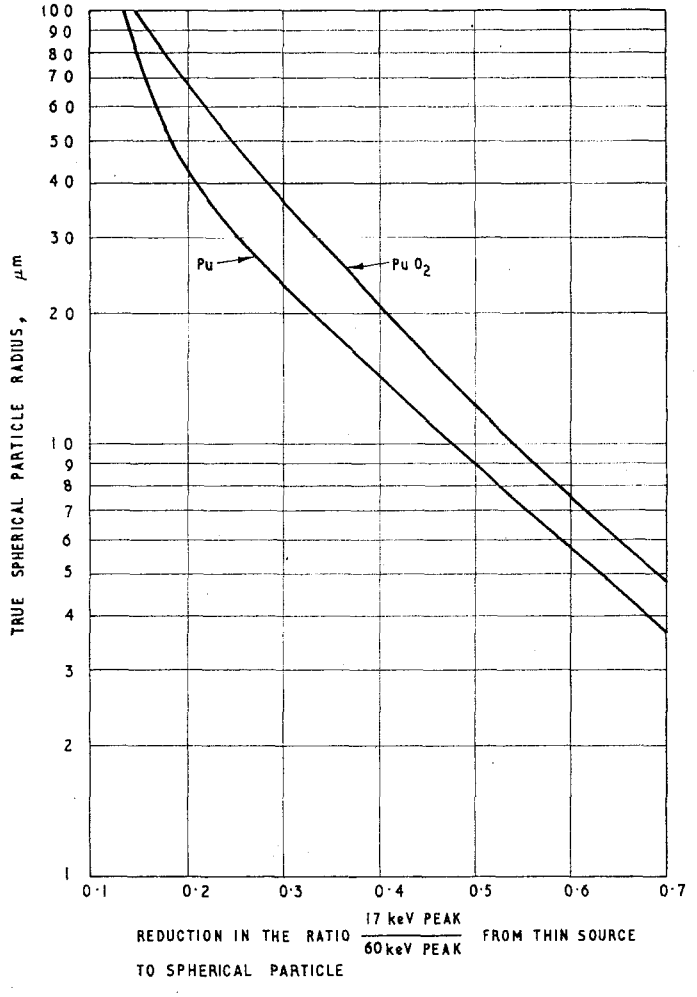

FIG. 6. Theoretical variation in the ratio $\frac{17 \text{ keV output}}{60 \text{ keV output}}$ with particle size.

The activity was then located as accurately as possible using the Monitor Type PNI 1080 with a number of masking devices, and the area was excised until monitoring showed that the contamination had been almost entirely removed.

As much as possible of the excised material, together with swabs, etc. was collected and analysed for plutonium. This measurement showed that at least 1·7 μCi of α activity had been removed from the wound. Monitoring the area of the wound then gave an estimated apparent residual alpha activity of about 0·005 μCi.

8.2. *Second Measurement*

After a further period of 10 months the finger was examined again, this time with a gamma spectrometer comprising a probe Type 1320X coupled to a multichannel analyser. The spectrum obtained is shown in Fig. 7. The ratio $\frac{17 \text{ keV peak}}{60 \text{ keV peak}}$ for this spectrum was very low

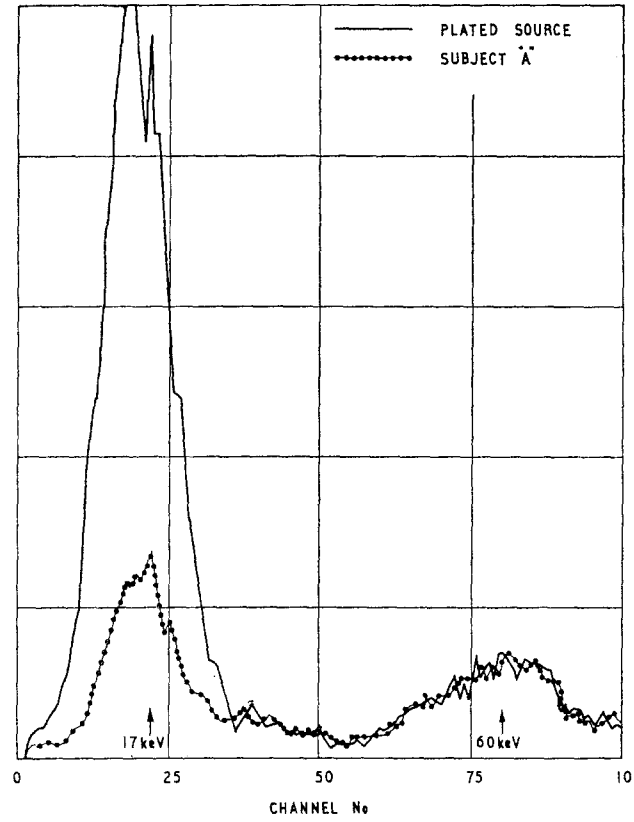

FIG. 7. Spectra from plutonium sources.

in comparison with that for a standard ^{239}Pu source which was used to set up the spectrometer and it was at first suspected that the plutonium in the wound must contain a high percentage of ^{241}Am. A thin electro-deposited source was therefore prepared from some of the material originally excised from "A"s finger. This source was calibrated in an alpha spectrometer giving an alpha activity of 0·023 μCi of which about 3% could be attributed to ^{241}Am. It was then used as a reference source for all future measurements on "A"s finger. The spectrum from the reference source is shown in Fig. 7. It has a much higher 17 keV peak. Comparison of the count-rates in the 17 keV X-ray region yielded an α activity in the finger of about 0·006 μCi, in good agreement with the original estimate. However a comparison at 60 keV indicated 0·022 μCi α activity in the finger. Applying Tyler's correction data for self-absorption in a single spherical particle of plutonium metal to the value at 17 keV, yields 0·020 μCi α activity in agreement with the 60 keV value. The estimate of alpha activity remaining at the wound site was therefore amended to 0·02 μCi approximately. This assessment made no allowance for the self absorption of 60 keV gamma rays nor for the possible effect of depth in the wound.

8.3. *Third Measurement*

The latest measurement on this subject was made 18 months after the original incident, using a similar probe coupled to a multi-channel analyser. Again the spectrum from the wound, after 20 min counting, showed 17 keV and 60 keV peaks and was compared with the spectrum from the reference source.

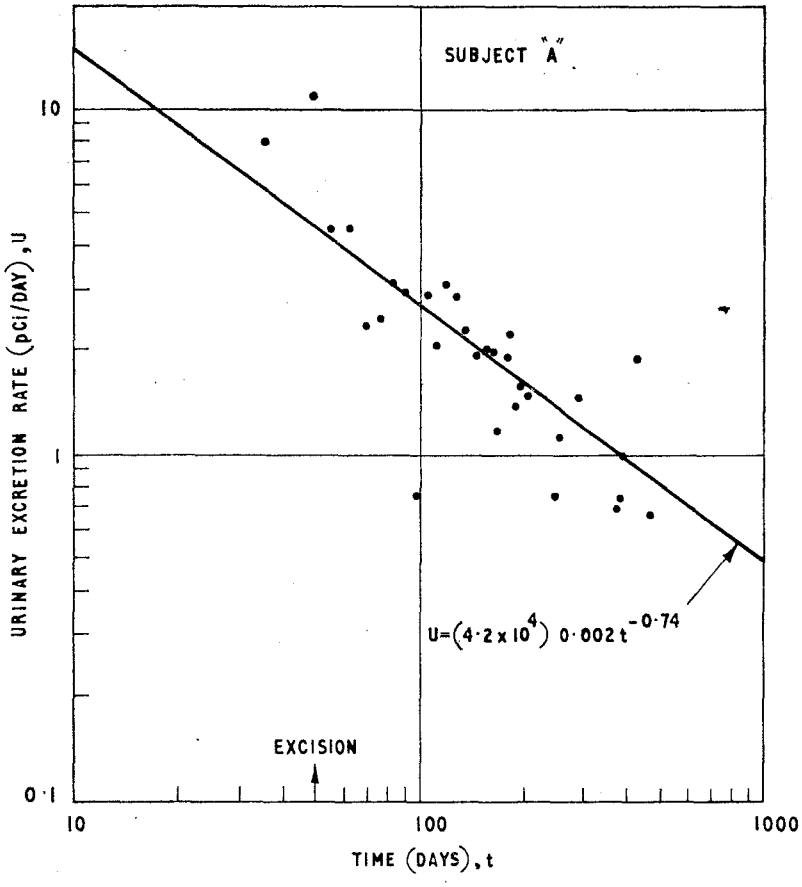

FIG. 8. Urinary excretion data for subject "A".

The total count at 17 keV corresponded to an apparent alpha activity of 0·004 μCi while that at 60 keV indicated 0·023 μCi.

The ratio $\frac{17\text{ keV}}{60\text{ keV}}$ was measured for each spectrum by integrating the total counts under the peaks. The values obtained were:

Electro-deposited reference source:

$$\frac{17\text{ keV peak}}{60\text{ keV peak}} = 6{\cdot}8$$

Wound contamination:

$$\frac{17\text{ keV peak}}{60\text{ keV peak}} = 1{\cdot}45.$$

This reduction in the ratio, if attributed solely to self-absorption in a particle of plutonium, corresponds to a particle radius of 30 μm (Fig. 6). However, such a particle would have an alpha activity of 0·12 μCi and would have given much higher total count-rates. Some of the reduction may be due to depth in the wound. In order to estimate this effect, spectra were plotted for the reference source covered by successive layers of tissue-equivalent plastic material. With 3 mm of plastic overlaying the source, the ratio $\frac{17\text{ keV peak}}{60\text{ keV peak}}$ was 4.9.

The reduction in ratio in going from the covered reference source to the actual wound now corresponds to a particle radius of 20 μm (0·035 μCi). The wound activity calculated from the total counts at 17 keV and corrected for self-absorption corresponds to a particle radius of 17·5 μm (0·024 μCi). The wound activity calculated from the total

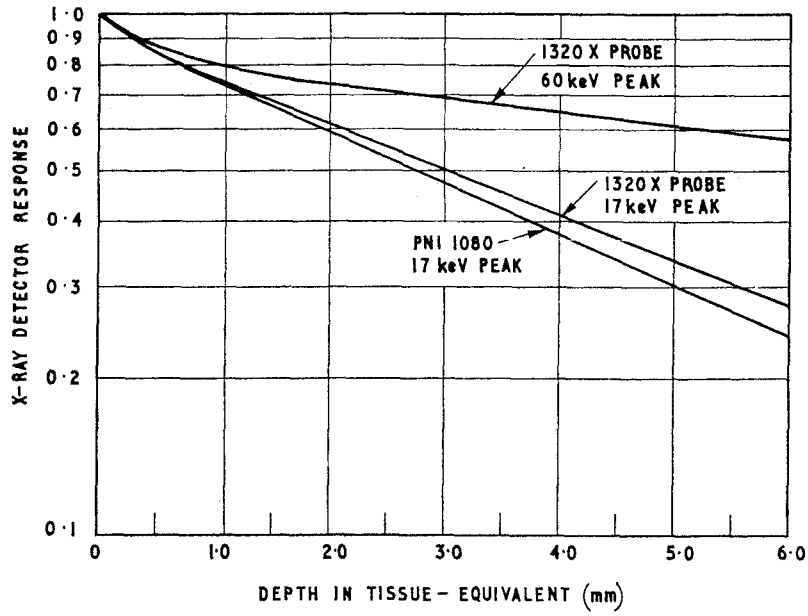

FIG. 9. Effect of depth in tissue on response of probes Type 1320X and PNI 1080 to radiation from thin source of plutonium.

counts at 60 keV and corrected for self-absorption corresponds to a particle radius of 18·5 μm (0·028 μCi). These are in fairly good agreement and it thus appears that the spectrum shapes and total counts are consistent with a plutonium particle of about 0·03 μCi alpha activity at a depth of about 3 mm below the surface.

9. GENERAL OBSERVATIONS CONCERNING THE ABOVE CASE

9.1. Monitoring with an alpha probe failed to detect the presence of plutonium in the finger. Simple monitoring with an X-ray probe after excision probably underestimated the residual activity by a factor of about 6.

9.2. The activity at the site of the wound does not appear to have changed significantly over a period of more than 1 year.

9.3. The results of urine analysis are shown in Fig. 8. They are consistent with the pattern suggested by Langham[8] for a plutonium injection of about 0·04 μCi into the system at about the time of the incident.

10. CONCLUSIONS

10.1. In the monitoring of wounds for plutonium contamination, using X-ray probes, errors can arise from self-absorption, from absorption in overlying tissue and from the presence of isotopes other than ^{239}Pu. These errors can be reduced if a thin reference source can be made from the same contaminating material and if both the wound and the reference source are examined using a low energy gamma spectrometer.

10.2. An actual case has illustrated the needs for reliable initial monitoring, for precise location and for accurate final assessment.

11. ACKNOWLEDGEMENTS

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