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A New Approach for an Improved Short-time Procedure for Locating and Measuring Low-activity, Low-energy Transuranium Deposits in Wounds

Removal of a Four-month-old ^{239}Pu Deposit in a Puncture Wound at the Tip of the Left Middle Finger

O. Fromheim, L. Ohlenschläger, W. Rapp

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A NEW APPROACH FOR AN IMPROVED SHORT-TIME PROCEDURE FOR LOCATING AND MEASURING LOW-ACTIVITY, LOW-ENERGY TRANSURANIUM DEPOSITS IN WOUNDS

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Abstract

A NEW APPROACH FOR AN IMPROVED SHORT-TIME PROCEDURE FOR LOCATING AND MEASURING LOW-ACTIVITY, LOW-ENERGY TRANSURANIUM DEPOSITS IN WOUNDS.

The paper describes a system in use at the Medical Department of the "Gesellschaft für Kernforschung", Karlsruhe for locating and measuring the activity of transuranium deposits inside wounds and at the surface of wounds. The measuring system consists of a set of two NaI(Tl) scintillation detectors particularly designed to detect and locate low-energy X-rays, and a third NaI(Tl) detector for the energy range up to 1 MeV. All three detectors have nearly the same sensitivity of 1 nCi for a ^{239}Pu point source and a measuring time of 100 s, but their geometrical resolution is different being especially optimized in the wound probe for locating a radiating point deposit within 2 to 3 mm. The detectors are held by a flexible mechanical support which allows the surgeon to manipulate the detector in the wound until the maximum response of the pulses produced is found by audio inspection. The nuclide is identified as far as possible, and the source activity is quantitatively estimated, with the help of the displayed spectrum of a multichannel pulse-height analyser.

INTRODUCTION

The existing apparatus for detecting low-activity and low-energy deposits in wounds is not adequate for providing the surgeon with the necessary information about the kind of isotope, the amount of activity, and the precise location within the shortest time possible for him to be able to give the best medical treatment. As all known monitoring systems lack one or more properties considered vital for an optimum treatment, it was worth our while to develop a new detector system with better location-finding capabilities for detecting a radiating deposit both in a subcutaneous wound and at a surface wound. An injury in which transuranium isotopes enter the wound is a serious hazard to the patient and requires immediate action to prevent the harmful effects. This also means that the surgeon must be able to remove the radiating deposit with as little tissue as possible.

From the medical point of view an optimum detector system should have the following properties:

(1) Good sensitivity in order to detect less than one tenth of the maximum permissible body burden.
(2) Good spatial resolution to be able to locate a deposit in a volume of no more than approximately 30 mm^3 adjacent to the detector.

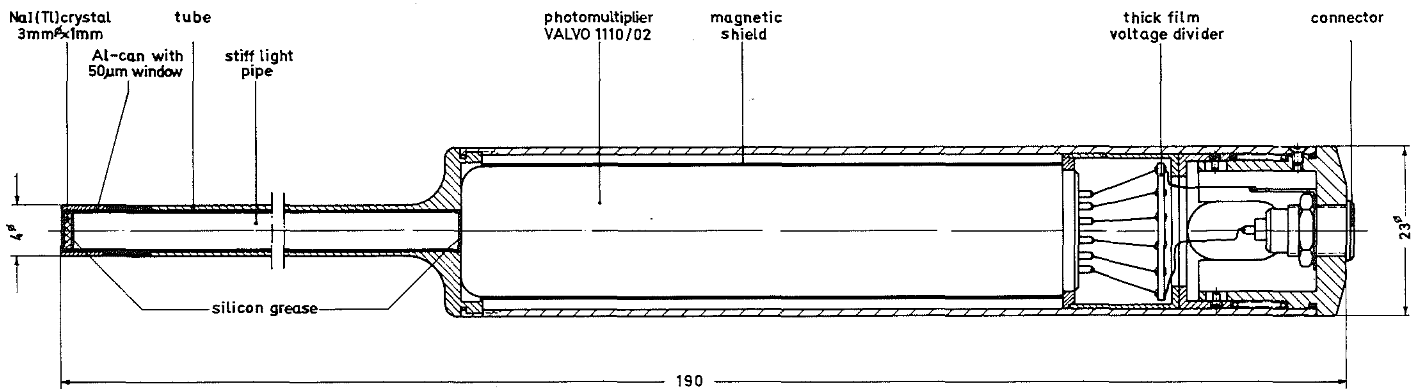

FIG. 1. Cross-section of the wound probe.

(3) Together with good sensitivity, a measuring time that is as short as possible in order to ease the discomfort of the patient who must nearly always endure a venous hyperaemia.
(4) Easy handling during the search for the radiating deposit and the possibility to be able to secure it in position when the deposit has been located. This leaves the surgeon with his hands free to work while he continues to survey the radiating point with proper hearing aids.
(5) The possibility to be sterilized.

On the basis of these requirements the question arises as to what kind of detector will provide the best results. Although semiconductor probes for radiation detection have been fabricated in the submillimetre range with excellent energy and spatial resolution, the choice was made in favour of a NaI(Tl) crystal probe with its inherent better absorption efficiency. Experiments with different semiconductor detectors proved to be unsatisfactory with respect to noise performance at body temperature and to counting time. If a thallium-activated sodium iodite scintillator is handled with the utmost care it can be used in a laboratory environment despite its hygroscopic sensitivity.

To ensure high counting efficiency the detector must be placed inside the wound. This circumvents the problem of mass absorption in tissue and improves considerably the geometric efficiency. The activity measurements are much more reliable when the detector surface is close to the radiating deposit. Particular consideration has been given to the detection of ^{239}Pu. As is well known from the literature, the decay of ^{239}Pu leads to excited states of ^{235}U which disintegrate mainly by internal conversion producing a small fraction of low-energy X-rays in the range of 13 to 21 keV. When the spectrum is measured with a NaI crystal it shows a maximum energy peak concentrated around 17 keV. The total number of X-rays produced is about 5 per 100 nuclear disintegrations. However, the emitted alpha-particles have only an average range of 40 μm, whereas the low-energy X-rays have a half-thickness in tissue of about 0.5 cm.

We found it very useful to develop at least two kinds of wound probes with equal sensitivity but with different spatial resolutions. This means that the searching procedure begins with one probe that inspects a large volume at the expense of spatial resolution and then, when the radiating volume has been detected, the other probe with the better spatial resolution is used until the maximum response is found.

THE WOUND PROBE

For ease and flexibility in handling, the detector is built in the form of a light-weight wound probe, which can be held in the hand, and a movable mechanical support. The wound probe has a small NaI(Tl) scintillation detector, 3 mm in diameter and 1 mm thick, mounted in a 6-mm-long aluminium can of 4 mm outer diameter (Fig.1). The crystal side of this can is covered with a 50 μm layer of aluminium foil. This part of the probe has caused many problems with light interference which has deteriorated the energy resolution. The aluminium can is cemented extremely carefully onto a 60-mm-long tube of nickel-coated brass which is attached to the part of the probe that can be held in the hand (the grip). Inside the tube a stiff light pipe transmits the produced photons to the light-sensitive cathode of a 20-mm-diameter photomultiplier which is housed in the grip. Careful selection of the silicone oil for optically coupling the crystal, light pipe and photomultiplier helps to improve the energy resolution. Thus, the crystal and light pipe can easily be separated from the grip if mechanical force should cause damage to the radiation window. Magnetic shielding of the photomultiplier is achieved with a foil of self-adhesive My metal.

To keep the wound probe as light and as small as possible the voltage divider system has been designed as a thick film resistance network. During the initial stage of development the preamplifier for cable matching was designed to fit into the grip, but experiments have shown that no signal deterioration of the energy resolution was observed so that only one cable of 2 m length was necessary for high voltage supply and signal transmission. Signal forming and amplification is done in a separate module. The total length has been reduced to 190 mm with a 60-mm front piece for convenient manipulation.

The fact that the probe can be fixed in any desired position within a few tenths of a millimetre from the wound has proven very helpful because the probe is still pointing at the radiating source while the surgeon starts to remove the piece of tissue in front of the detector.

Sterilization of the detector has been accomplished by first cleaning the probes with an antiseptic solution and then coating the slender cylindrical tube with a sterile colourless lacquer. The laquer gives a thin protective layer after dessication for 10 min and is stripped off the probe after each surgical operation. After the tip of the wound probe has been sterilized the grip is fastened in the mechanical support and covered with a sterile elastic hose-like gauze. As the performance of the monitoring system is routinely checked every day, calibration is not necessary after sterilization except for a short functional test with a known ^{241}Am source.

THE MONITORING SYSTEM

The monitoring system consists of several types of detector optimized for different detection situations. Besides detectors for low-energy X-ray measurements, an alpha-beta counter was integrated into the measurement system in order to scan the surface of the wound with Lucite collimators for any transuranium deposit. This is not a reliable test since blood or tissue fluid will stop the penetration of alpha particles.

The measurement system consists of a 256-channel analyser for determining the energy distribution which can be displayed on a screen plotted on an X-Y plotter or registered on a printer within selectable energy ranges. As there is no time in an emergency for setting up instrumentation or for making calibrations, the monitoring system must be ready for immediate use. Consequently, our system is controlled from a central control board which displays all the necessary functions for the technical handling of the instruments. By a simple finger touch on a sensor the desired program is initiated, and by a second touch the program is executed. We distinguish between four main programs:

(1) Alpha particle counting
(2) Measuring of spectograms with one out of three selectable NaI detectors

(3) Registration of the spectrum within selectable channels with a plotter or a printer
(4) Summation of selectable channels for activity determination

A measuring time of 100 s was the most suitable for obtaining the best overall sensitivity of the system and the desired result of the measurement needed for further quick surgical treatment. A display of counting time or channel number and channel contents proved very convenient for making a quick survey.

Locating ^{239}Pu deposits with activities of a few nanocuries is very time consuming and also very unreliable if the surgeon relies solely on visual observation of the produced pulses. Therefore an audio inspection of the statistically emitted pulses is absolutely essential. Audio inspection by means of earphones was found to be the best method for achieving optimum concentration during the process of locating the deposit. In searching for rare statistical events the human senses are often deceived and not only by background noise. To overcome this, an audio discriminator has been improved by weighting the pulse amplitudes with six different frequency modulations, each individually or in selectable groups.

The determination of unknown nuclides is very difficult, especially in an emergency where simplification must and can be accepted. A simple indication of the presence and location of a deposit is enough information to enable further medical steps to be taken. It has become a convenient and, in most cases, successful procedure to compare the measured spectrum with the spectra of known isotopes after surgical treatments.

THE X-RAY DETECTORS

Design criteria

To achieve a good signal-to-noise ratio the crystal thickness is selected so that the absorption efficiency is as high as possible and the background rate is as low as possible. For the detection of ^{239}Pu a 1-mm-thick NaI(Tl) is required to absorb 99.9% of the resulting maximum 17-keV X-rays. The detector window consists of a 50-μm layer of aluminium, which is optimal for the mechanical construction, but not optimal for the transmittancy because only 94% of the normally incident 17-keV X-rays reach the crystal and are subject to photon conversion. The stiff light pipe transmits the produced photons of about 0.42-μm wavelength with an efficiency of 85%. Optical losses in the coupling layers are minimized by using silicone oil with the proper fraction coefficient. The spectral quantum efficiency of the photomultiplier (S-11) is matched to the wavelength of the photons. The only part in the system that could not be optimized is the photosensitive area of the cathode. The unused area contributes undesired noise even when the photomultiplier is selected for minimum dark current.

Results

Figures 2 and 3 give examples of the performance of the wound probe. In order to specify the resolution we refer to the photopeak of ^{241}Am at 60 keV. The wound probe shows an energy resolution of 25% with a spatial

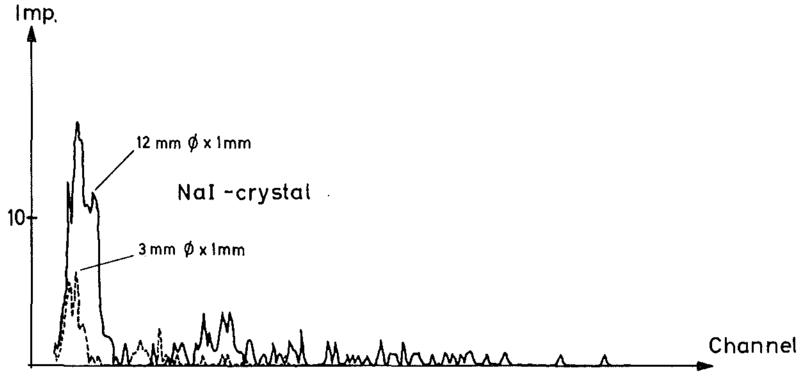

FIG. 2. ^{239}Pu spectrum of a 2.8-nCi point source from the wound probe. Counting time 100 s.

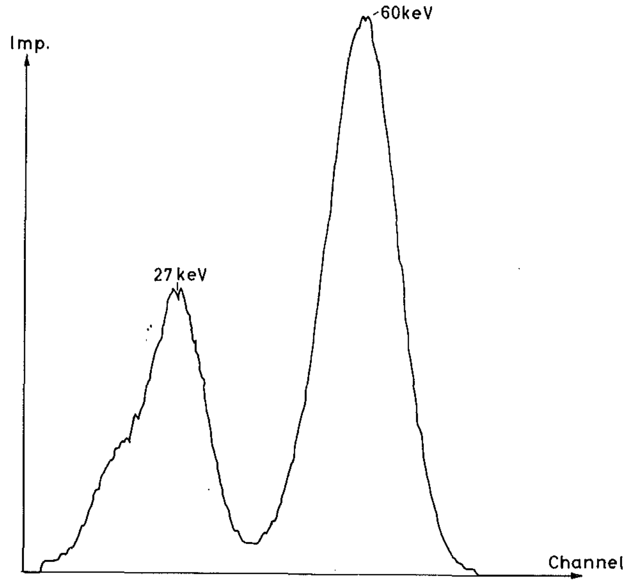

FIG. 3. ^{241}Am spectrum from the wound probe.

resolution of a volume of 30 mm^3 adjacent to the detector surface. Measurements with ^{239}Pu point sources have shown an activity as low as 1 nCi can be detected with a 17-keV channel count-rate equal to that of the background. In fact, the detector system, and especially the wound probe, were successfully used in animal experiments where it was possible to locate a deposit of 2 nCi of ^{239}Pu as a point source within a 1-cm-deep wound.

The plutonium sources used in the experiments had the following isotopic contents in per cent:

^{238}Pu	0.007
^{239}Pu	94.915
^{240}Pu	4.893
^{241}Pu	0.174
^{242}Pu	0.011

The good spatial resolution of the wound probe is due to its small diameter with the range-limited geometrical efficiency. Outside a distance equal to its diameter the efficiency drops to about 10% of the value at the surface of the detector. This means that once the maximum response of a low-energy deposit has been detected the unknown material must lie in the volume adjacent to the detector surface. Outside this range the wound probe is very insensitive. To ensure good sensitivity for the detection of low-energy deposits two more detectors with different crystal diameters have been added to the instrumentation. A second wound probe 6 mm in diameter completes the set for the direct wound measurements. A detector with a 12-mm-dia. and 1-mm-thick crystal is intended for application on wound surfaces. This detector can be replaced by one with a 12-mm-thick crystal to extend the energy range up to 1 MeV.

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DISCUSSION

G.B. SCHOFIELD: I think the probes you showed in the film following your presentation represent a most useful addition to the plutonium detection systems used in wound assessment. In practice, of course, most wounds are too small to allow deep penetration of the probe into the wound. In large wounds, however, and in particular where diffusion occurs, a small probe of this type could be really useful. The instrument would also be very valuable in the operating theatre for continuous monitoring of the operation site and for monitoring small pieces of excised tissue to check whether the radioactivity has been removed.

T.A. LINCOLN: In my limited experience with contaminated wounds, especially puncture wounds, the contaminant is frequently either a liquid or a particulate material. Do you have any trouble with radioactive contamination of the tip of the probe itself? How do you decontaminate the probe during exploration and treatment of a wound?

O. FROMHEIN: We had no problems at all in cleaning the tip of the probe during the wound examinations. When we experimented with fluid radioactive material the tip was eaisly cleaned using a piece of cloth wetted with toluol.

D. RAMSDEN: In your standardization procedure do you allow for absorption of the 17-keV radiations in the contaminating body, i.e. a plutonium-oxide particle or, in your case, the glass?

You quote an estimated activity of 3 nCi by external detection. What activity was found in the excised fragments?

O. FROMHEIN: The glass pipette was radioactive waste and was contaminated all over its surface. When the deposit had been located, as indicated by the maximum acoustic response, the contaminated area in front of the detector was excised. A second wound measurement showed zero-effect, which meant that all the radioactive material had been found. The amount of activity was estimated by comparison with a known ^{239}Pu source and was found to be less than 5 μCi. The result was checked by radiochemical analysis of the excised tissue and of the glass splinter, and a figure of 2.8 μCi was obtained.

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REMOVAL OF A FOUR-MONTH-OLD ^{239}Pu DEPOSIT IN A PUNCTURE WOUND AT THE TIP OF THE LEFT MIDDLE FINGER

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Abstract

REMOVAL OF A FOUR-MONTH-OLD ^{239}Pu DEPOSIT IN A PUNCTURE WOUND AT THE TIP OF THE LEFT MIDDLE FINGER.

The necessary diagnostic and therapeutic measures taken in a case of a 4-month-old plutonium-239 wound deposit are discussed. The development of a wound probe detector allows deposition of transuranium elements in wounds to be measured and, consequently, gives a better location of the wound activity, thus providing the possibility of selective and careful removal of the wound deposit. The detection limit of the wound probe is about 2 nCi. The results of the measurements with the wound probe are discussed for the case of a 4-month-old plutonium-239 deposit in a puncture wound on a finger tip.

INTRODUCTION

A wound contaminated by highly radiotoxic transuranium elements needs special medical care with respect to the diagnostic and therapeutic measures to be taken because of the potential danger to the whole organism. The radionuclide moves via opened lymph and blood vessels into the organs where its deposition becomes a potential internal radiation hazard.

The radioactive material can enter the wound as a solute, a soluble solid or an insoluble solid. With the soluble and soluble solid forms there is a risk of fast incorporation through opened lymph spaces and blood vessels. Wound contaminations by insoluble transuranium elements lead to a stationary wound deposit with a high local radiation burden. If the wound is contaminated by soluble plutonium, it moves into the blood stream at a relatively quick rate and part of the isotope is eliminated via the kidneys. The largest fraction is carried, partly by endogenic complexing, to the organs and deposited. Plutonium-239 is preferentially deposited in the glycoproteins and mucoproteins of the endosteal and periosteal surfaces of the bone trabeculae [1]. In addition, it is deposited in the liver. Only a small fraction of the plutonium deposited in the organs is mobilized and eliminated via the kidneys and intestinal tract. On account of the very long biological half-life of about 100 years, the organism is exposed to continuous internal radiation which can lead to serious damage to the health. The amount of plutonium that can cause these potential lesions is extremely small. The maximum permissible amount of plutonium in the body has been fixed at 40 nCi = 0.6 μg by the International Commission on Radiological Protection [2]. Maximum permissible values for wound contamination are not available. However, according to a rule of thumb the plutonium wound activity should not exceed 1/10 of the maximum permissible body burden because of the high local radiation exposure.

RADIATION MEASUREMENT TECHNOLOGY FOR A PLUTONIUM-CONTAMINATED WOUND

An essential prerequisite for promissory treatment of a contaminated wound is the determination of the level, nature and extent of the wound activity. This calls for sensitive measuring equipment to enable the attending physician to record even the smallest activity. This is particularly true for wound contaminations caused by highly radiotoxic transuranium elements. While X-rays, gamma rays and high-energy beta radiation can be recorded with certainty by the measuring techniques because of the greater range they cover in the tissue, it is extremely difficult to detect alpha radiation either on or within the contaminated wound because of the radiation absorbed by haemorrhage, blood coagulation and scab formation. Consequently, to obtain an accurate recording of the wound activity when transuranium elements are present in a wound deposit an additional measurement has to be made of the low-energy X- and gamma rays. This must be accomplished by the measurement procedure carried out at the site of the contaminated wound and within the wound. First, the alpha radiation is measured by means of a large-area proportional counter. To rule out any deeply situated wound deposit, the low-energy X- and gamma radiation are measured at the wound [3]. Then the wound probe detects the exact location of the wound deposit by measuring the X- and gamma rays within the wound. A minimum detection sensitivity of 4 nCi is required for a plutonium wound deposit. As soon as the diagnostic measurements have been made the medical therapy can begin.

THERAPEUTICAL MEASURES

The aim of the therapy is to remove with the utmost care the entire wound deposit. The prerequisites for the surgical treatment of a contaminated wound are an appropriate room, adequate measuring techniques and suitable equipment. To avoid contaminating a conventional operating theatre with radioactivity, this type of injury should be treated in a room specially designed for the purpose. All the instruments, protective sheets, side-tables, operation gloves etc., should be used exclusively for the treatment of contaminated wounds.

If the cutaneous areas around the wound are contaminated, these must first be removed in order to avoid spreading the contamination in the operating theatre.

The surgical procedure is determined by the nature and severity of the lesion and by the radiotoxicity of the radioactive material present in the wound.

Conservative surgical treatment is sufficient for wounds contaminated by short-lived radionuclides. At its simplest this entails wound irrigation under running water. In addition, the wound can be washed with physiological saline or Zephirol water.

Active surgical measures are preferred for all wounds contaminated by highly toxic substances including the long-lived bone-seekers. Most of these lesions are located on the fingers and hands as a result of work performed in glove boxes. In those cases where dissolved radioactive substances are involved and a venous stoppage has not been applied in the vicinity of the wound, diffusion of the substance into the blood circulation must be anticipated. Therefore, the first step to take is to apply a venous stoppage and then measure the wound activity. Next, the wound edge is excised under regional or local anesthesia. Experience has shown that in most cases the total activity can be removed from the wound in this way. The excised tissue, the wound and venous blood as well as the washings and first-aid dressings should all be measured for activity. If diffusion and incorporation of the radioactive material are suspected, activity measurements must also be made on samples of blood, urine and faeces. An additional therapy with medicaments must be considered whenever a substantial incorporation is suspected. In severe "combined" lesions, such as contamination accompanied by a severe general trauma, the primary measures taken will depend on the criteria of first-aid treatment. The therapy for severe conventional wounds has priority for the purpose of saving life.

A FOUR-MONTH-OLD WOUND DEPOSIT IN A PUNCTURE WOUND AT THE TIP OF THE LEFT MIDDLE FINGER

Case history

While disposing of radioactive waste at a nuclear research centre operated by the European Community an employee punctured the tip of his left middle finger four months before he was transferred to the Medical Department of the Karlsruhe Nuclear Research Center. Since the worker had kept the accident secret, immediate treatment of the wound was not possible. Meanwhile, the wound had healed without showing signs of irritation. Routine incorporation monitoring two months after the incident showed an increase in the plutonium activity in the urine. The worker was questioned about this increase and information was obtained about the finger injury. An inhalation accident was ruled out both on grounds of anamnesis and faeces analyses. The activity measurement of the tip of the left middle finger that had been made elsewhere recorded about 2.8 nCi of plutonium-239. The patient was admitted to the Medical Department of the Karlsruhe Nuclear Research Center for further medical treatment.

Findings

The tip of the left middle finger appeared to be normal and there were no detectable signs of an earlier lesion. As expected, measurement of the left hand, including the tip of the left middle finger, by means of a large-area proportional alpha counter gave a counting rate of zero but measurement of the low-energy X- and gamma radiation at the tip of the left middle finger by a scintillation detector confirmed the plutonium deposit of some 3 nCi already measured elsewhere.

Results of measurement with the wound probe detector

Under regional anesthesia and using an Esmarch's bandage the tip of the left middle finger was incised in the region of measured activity and

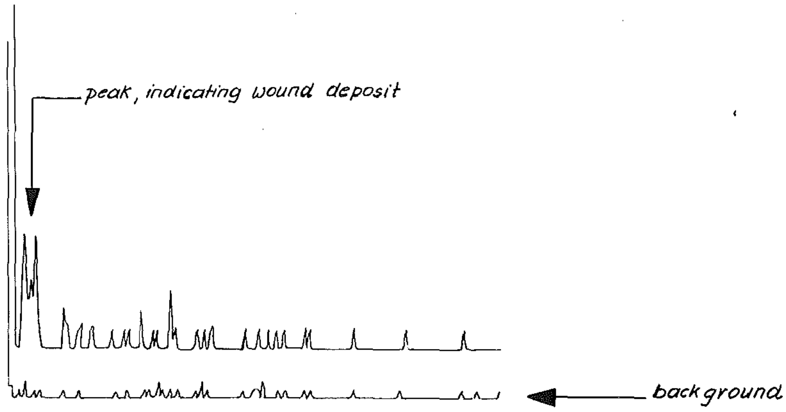

FIG.1. Measurement of four-month-old ^{239}Pu wound deposit made within the wound.

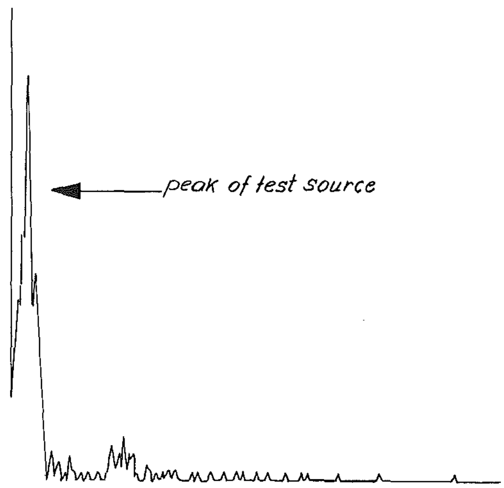

FIG.2. Measurement of ^{239}Pu test source.

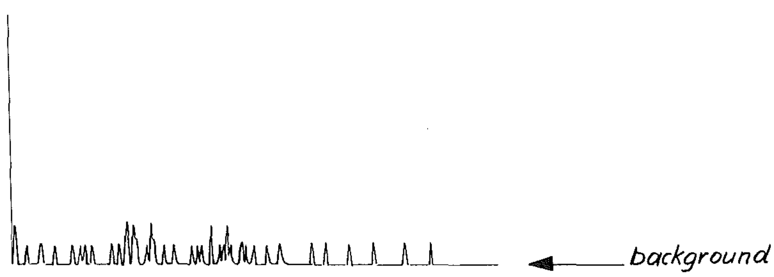

FIG.3. Wound monitoring after excision of the ^{239}Pu deposit.

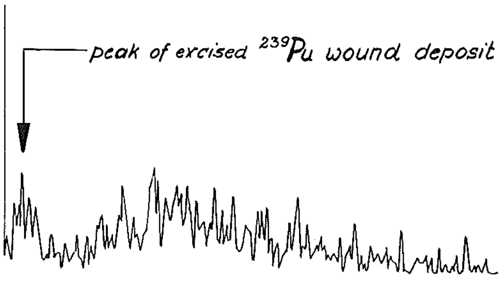

FIG.4. Check measurement of the excised tissue.

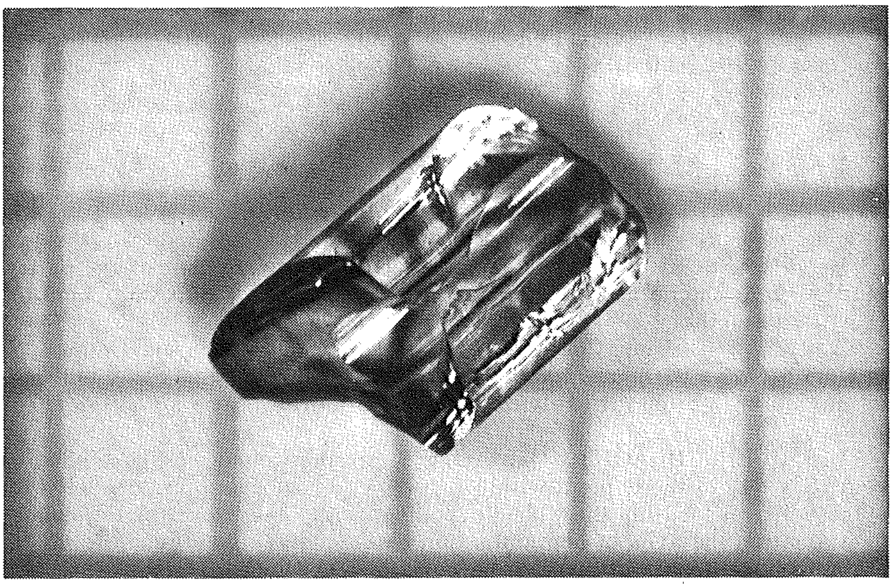

FIG.5. Broken tip of a plutonium pipette found in the excised tissue.

the wound deposit was located precisely by carefully guiding the probe into the wound (Fig. 1). A test measurement was made with a plutonium preparation of known activity to support the diagnosis (Fig. 2). Then the plutonium wound deposit was removed selectively. Measurements made with the wound probe in the wound after the deposit had been excised confirmed that the activity had been removed (Fig. 3). The excised tissue containing the activity was measured again with the scintillation counter to reproduce the measured result (Fig. 4). The radiation source detected in the excised tissue was an approximately 2-mm-long plutonium-contaminated glass splinter from a plutonium pipette (Fig. 5).

ACKNOWLEDGEMENTS

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DISCUSSION

E.S. WILLIAMS: Considering the likelihood of diffusion from the site, did you continue to monitor the patient's urine and, if so, with what result?

L. OHLENSCHLÄGER: After excision of the wound deposit the patient returned to his home country. Several months later we were informed that the patient's urinary excretion had decreased.

A. LINDENBAUM: Could you give us some idea of the maximum distance between the probe and the radioactive source. I ask this because the plutonium is often diffusely deposited, or there may be some intervening tissue.

L. OHLENSCHLÄGER: Measuring within the wound the probe should accurately detect the deposit within a distance of about 2 mm. An acoustic device helps in locating the wound deposit. In the case of diffuse wound contamination the area of contamination can be determined by using the four detectors. Residual activity can be measured and located with the wound probe. The excised tissue should always be measured to judge the success of the operation.

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