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Part 3

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PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1969
TO THE
USAEC DIVISION OF BIOLOGY AND MEDICINE
VOLUME II: PHYSICAL SCIENCES
PART 3. INSTRUMENTATION

March 1970

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DEVELOPMENT REPORT

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PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1969
to the
USAEC Division of Biology and Medicine
VOLUME II: PHYSICAL SCIENCES
Part 3. Instrumentation

by

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and
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and
Electro-Optical and Sonics Group

March, 1970

BATTELLE MEMORIAL INSTITUTE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

Volume I of this report to the USAEC Division of Biology and Medicine covering work in the Biological and Ecological Sciences, the latter with contributions from the Water and Land Resources Department, is issued as BNWL-1306, Parts 1 and 2, respectively. Volume II of this report covers work in the physical sciences and is issued in three parts: Atmospheric Sciences, BNWL-1307, Part 1; Radiological Sciences, BNWL-1307, Part 2; and Instrumentation, BNWL-1307, Part 3.

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FOREWORD

The Radiation Instrumentation part of the program focuses on the development and evaluation of advanced instrumentation concepts, detectors, and circuitry, and on their feasibility in the detection and measurement of ionizing radiation and other physical quantities of interest to Division of Biology and Medicine programs. The present emphasis is on equipment for the measurement in vivo of plutonium depositions.

A portion of the effort is directed to cooperative research and development with other Division of Biology and Medicine programs at the Pacific Northwest Laboratory involving new detection methods and instrumentation for application to biological, dosimetric, and environmental research.

ORGANIZATION

These reports are drawn principally from personnel of the Bio-Medical Instrumentation Group of the Applied Physics Department.

The following organization chart depicts the relationship to the Pacific Northwest Laboratory of the investigators involved with the instrumentation program for the Division of Biology and Medicine.

Systems and Electronics Division, C. A. Bennett, Manager

PREVIOUS REPORTS IN THIS SERIES

HW-70050	Period Ending December 31, 1960
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X RAY TO ALPHA RATIO STUDIES

K. L. Swinth

Both experimental and computational studies of the X ray-to-alpha ratio (X/α) were performed for plutonium isotopes. Using measured X/α ratios for pure isotopes and the known isotopic composition to predict X/α ratios for plutonium isotopic mixtures, good agreement with measured results was obtained. The calculated absorbed dose rate was found to increase with decreasing ^{239}Pu content (i.e. higher burn-up) in a plutonium isotopic mixture and also to increase with the growth of ^{241}Am in that mixture. On the other hand, it was found that the calculated maximum permissible body burden per unit weight decreased with the growth of ^{241}Am and increased with decreasing ^{239}Pu content.

Measurements of X/α ratios were extended to ^{244}Cm and ^{242}Cm yielding values of 0.093 and 0.097, respectively. In the case of ^{242}Cm corrections were required for decay and

for the contribution from ^{244}Cm and ^{241}Am in the material. The solid state counter described in the next paragraph should reduce this uncertainty due to its ability to resolve the alpha particles from the various isotopes.

An all solid-state X-ray-to-alpha coincidence counter was developed, and preliminary results indicate an X/α ratio for ^{239}Pu to be about 4% lower than that previously obtained with the scintillation detector NaI(Tl)-proportional counter unit. The solid-state system uses a coaxial geometry with the alpha detector sandwiched between the source and X-ray detector. A 3 mm $\times$ 110 mm² lithium-drifted silicon detector cooled to 77 °K is used for X-ray detection, while a totally depleted surface barrier detector (40 μ $\times$ 100 mm²) detects the alpha particles. Sources are electrodeposited on 7/8-in. diam stainless steel discs positioned 1.13 cm from the

X-ray detector and 0.45 cm from the alpha detector. A rotatable disc located inside the vacuum chamber holds six sources.

The X-ray detector has a resolution of 1.05 keV FWHM at 26.4 keV and an average efficiency of 3.6% for the X rays from ^{241}Am . The alpha detector resolution is 78 keV FWHM and its efficiency is 14.6% for alphas from ^{241}Am . These efficiencies nearly double when a point

source is used. A previous solid-state system, with efficiencies almost an order of magnitude lower, created severe source handling problems. The NaI(Tl)-proportional counter system, on the other hand, has efficiencies about five times better than the solid-state system, but lacks the resolution, therefore making data interpretation more difficult.

Neg 700517-6

Geometrical Arrangement of Source and Detectors Within the Solid-State X Ray-to-Alpha Counter. The relation of the other major physical components is also illustrated.

Neg 700517-1

Comparison of Calculated (Solid Lines) and Measured (Open Circles) X Ray-to-Alpha Ratios for Two Plutonium Isotopes Showing Changes Occurring with Time. The good agreement between calculated and measured values is evident on this figure.

Neg 700517-7

Relative Detection Efficiency of X Rays in the all Solid-State X-Ray-to-Alpha Counter. The effect of the alpha detector in attenuating the X rays is shown on the figure. It causes the peak efficiency at about 19 keV.

PLUTONIUM WOUND COUNTING

K. L. Swinth and F. R. Reich

Investigations in the area of plutonium wound counting have centered on the application of surface-contoured diodes and on the development of three dimensional wound viewing techniques.

Surface-contoured diodes provide a small maneuverable detector for work close to a wound, but suffer from temperature instability and a low intrinsic counting efficiency (5%). Measurements of the attenuation of plutonium X rays in tissue-equivalent plastic show greater attenuation when using a surface-contoured detector as opposed to a sodium iodide scintillation counter. This is because the contoured diode responds with greater efficiency to the lower energy L X rays from the decay of plutonium. The diode attenuation data can be resolved into two components showing attenuation coefficients of about 1.8 cm^{-1} and 1.0 cm^{-1} . This compares favorably with coefficients of 2.2 cm^{-1} and 1.2 cm^{-1} for water at 13.6 keV and 17.0 keV respectively: the energies of two of the L X rays.

Measurement of the change in sensitivity with voltage applied to a surface-contoured diode showed a coefficient of 1.3% V. With a

change of 2.13 V/°C in the avalanche voltage, this indicates a change of 2.8%/°C in the sensitivity for the L X rays. Again, this shows the need to control the temperature environment of the detector or in other ways compensate for temperature drifts. Background measurements indicate a background rate of 0.1 cpm. A series of 100 consecutive counts of 30 sec was zero, except for five with one count each.

In complicated wounds, direct visualization of the distribution of material within the wound area is desirable. If a three dimensional view or map of the wound can be formed, erroneous or unnecessary removal of tissue can be avoided. This led to consideration of an image intensifier system with a three-dimensional readout. Theoretical considerations show the feasibility of such a system built around a low-noise image intensifier. In order to study the three dimensional imaging characteristics, a simulated wound distribution was created and imaged on film. Holograms were formed and yielded a good three-dimensional representation, but did not appear to be any better than a simple stereo-pair.

Neg 700517-4

Attenuation by Tissue of ^{239}Pu X Rays Measured by a Surface-Contoured Diode. Similar data taken with a NaI(Tl) scintillation detector are shown.

Neg 700517-3

Change in Counting Efficiency Caused by a Change in the
High Voltage Applied to a Surface-Contoured Diode Detector.

INSTRUMENTATION FOR IN-VIVO PLUTONIUM COUNTING

K. L. Swinth

Studies on phoswich detectors, low-noise phototubes, and phototube noise reduction techniques were carried out in the development of instrumentation for the lung counting of plutonium. In order to test the phoswich detectors and phototube noise reduction techniques, an electronic pulse-shape discrimination system using a time to pulse height converter was established. A leading edge discriminator is used to detect the start of a pulse, and a zero crossing discriminator detects the zero crossing of the doubly differentiated pulse. A time to pulse height converter measures the time between these events which shifts with changes in pulse risetime.

Many techniques of risetime discrimination were considered, but coincidence with the pulse risetimes of interest, as measured by a time to pulse height converter, gave as good results as the other techniques tested. Excellent discrimination between pulses with risetimes in the neighborhood of 1 nsec, 250 nsec and 1.1 μ sec was obtained over the energy range of interest (10 keV to 100 keV).

Use of the pulse risetime discrimination technique on a sodium iodide-cesium iodide phoswich detector yielded background rates of 0.012 cpm/cm² in the energy range from 13.6 to 25.6 keV. The detector is a 3 mm by 5 in. diameter sodium iodide crystal

Shift in Time to Pulse Height Converter Output Level With Changes in Risetime and Pulse Height. The peak channel and approximate spread at half height are given.

Risetime, nsec	Peak Channel for Pulse of Energy Equivalent to:		
	15 keV	30 keV	60 keV
100	13 $\pm$ 3	15 $\pm$ 1	17 $\pm$ 1
150	17 $\pm$ 3	20 $\pm$ 1	21 $\pm$ 1
200	29 $\pm$ 4	33 $\pm$ 2	33 $\pm$ 1
1000	110 $\pm$ 6	118 $\pm$ 5	123 $\pm$ 2

backed by a 2 in. cesium iodide anti-coincidence detector. Output pulses were gated in coincidence with the sodium iodide pulses giving a background reduction of almost two orders of magnitude over the ungated output. The background rate of 0.012 cpm/cm^2 is only one sixth the background rate of 0.069 cpm/cm^2 obtained using a large area of (805 cm^2) 1 mm thick by 1-3/4 in. NaI(Tl) crystals on low-noise 2-in. phototubes. By surrounding the large area counter with plastic phosphor a similar background reduction (to 1/7) was obtained.

Noise pulses from phototubes extend into the energy region of interest for plutonium counting. Pulse rise time discrimination can be used to gate out these fast noise pulses. Elimination of noise pulses in the X-ray region (13.6 to 25.6 keV) reduced the background by 26% in

the large-area (805 cm^2) thin-scintillator counter. This represents a dark noise background rate of about 0.02 cpm/cm^2 over the photocathode area. Tests on a larger crystal (2 mm $\times$ 5 in. diameter) and low-noise photomultiplier (5 in.) yielded a reduction of 40%.

Tests were performed on a number of off-the-shelf phototubes to evaluate their use in counting the uranium L X rays resulting from the decay of plutonium. In the energy range from 13.6 to 25.6 keV, the dark noise background rate varied from 0.015 cpm/cm^2 to 0.034 cpm/cm^2 for the 2-in. tubes that were tested. A tube with a bialkali photocathode fell in the middle of the range, but was judged to be the most promising because of increased resolution. It had a full-width at half maximum of 52% as opposed to typically 65% for the others.

Neg 700517-5

Block Diagram Showing the Scheme Used in Pulse-Shape (Risetime) Discrimination Experiments with Scintillation Detectors.

A SOLID-STATE RADON MONITOR*

K. L. Swinth

In order to monitor radon daughter levels in a facility designed to expose small animals to controlled levels of radon and radon daughters, a simple counting system was built based on alpha spectroscopy. The unit built utilizes commercially available modules and a surface-barrier detector. Filters, with the deposited activity taken from an air sampling head positioned in the exposure chamber, are placed in the counter, where, at selected intervals after sampling, the alpha activities of RaA and RaC' are determined. The sequence of sampling and counting is controlled by a timer to maintain ease of operation and accuracy. One standard AEC instrument bin houses the entire instrument minus the sampling head.

From two sets of counts, the concentrations of RaA, RaB, and RaC in the sampled volume can be determined. Equations were developed permitting a simplified graphical analysis of the results. However, a more rigorous calculation can be used for improved accuracy. The spectroscopy

method uses calculations similar to the Tsivoglou method, but is simpler and more accurate than the Tsivoglou method. When ratios of counts are used, simplified calculations can be performed that permit graphical analysis. These equations are illustrated below.

$$\text{RaA} = K_1 C_{11}$$

$$\text{RaB/RaA} = K_2 \frac{C_{32}}{C_{11}} - K_3 \frac{C_{31}}{C_{11}} + K_4$$

$$\text{RaC/RaA} = K_5 \frac{C_{31}}{C_{11}} - K_6 \frac{C_{32}}{C_{11}} + K_7$$

where:

C_{11} = RaA counts during first counting interval

C_{31} = RaC' counts during first counting interval

C_{32} = RaC' counts during second counting interval

The K's represent constants unique to the parameters of the counting system and the decay characteristics of radon and its daughters. These constants vary with the counting efficiency, the sampling time, and the time and duration of the counts taken after sampling.

* This activity was funded through the Inhalation Toxicology Section, Biology Department

The counter has a detection efficiency of 0.201 ± 0.006 , and measurements show that degradation of the alpha spectrum in the filters is not a problem. Radon levels in the sampled chambers are on the order of 7 nCi/liter and 100 nCi/liter. In one chamber, a sequence of samples

following equilibrium yielded a RaA concentration of 69.4 ± 3.7 nCi/liter. The measured ratio of RaA:RaB:RaC was 1:0.25:0.14. A separate semi-continuous system is used to monitor the radon levels in the chambers. Collected data have been compared with radon and radon daughter levels analyzed by other methods.

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R. L. Wilbur. "Instrument for the Measurement of Surface Tensions of Aqueous Surfactant Solutions," Review of Scientific Instruments, Vol. 40, p. 355. 1969

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OPEN LITERATURE PUBLICATIONS

C. E. Cushing and N. S. Porter. "Radionuclide Cycling by Periphyton: An Apparatus for Continuous in situ Measurements and Initial Data on ⁶⁵Zn Cycling," in Symposium on Radioecology, CONF-67-503. Proceedings of 2nd National Symposium, Ann Arbor, Michigan, May 15-17, 1967

PRESENTATIONS AT SOCIETY MEETINGS AND SYMPOSIA

K. L. Swinth. "In Vivo Plutonium Counting Facilities and Research at Battelle," Fourteenth Annual Meeting of the Health Physics Society, Pittsburgh, Pennsylvania, June 8-12, 1969.

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