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Biomedical probe using a fiber-optic coupled scintillator*

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A high-sensitivity biomedical radiation probe which employs a fiber-optic coupled NaI(Tl) scintillator as a detector is described. It was developed for *in vivo* counting of low-energy ^{239}Pu photons from material located in the tracheobronchial lymph nodes. This probe is 20 times as sensitive as a solid-state probe (avalanche diode) previously developed for this application. Tests with ^{99m}Tc show a sensitivity more than 90 times greater than biomedical probes using DcTe of GaAs; however, the improved sensitivity is largely due to an increased sensitive volume. Probes were evaluated in animals and phantoms for detection of ^{239}Pu and for location of lung tumors labeled with ^{111}In.

I. INTRODUCTION

A biomedical scintillation counter similar in size to a fiber-optic bronchoscope was built to count low concentrations of internal radioactivity. The probe was built specifically to count the uranium L x rays (average energy 17 keV) emitted in the decay of plutonium located in the tracheobronchial lymph nodes of accidentally exposed plutonium workers.[1] The lymph nodes are accessible for counting by placing a radiation detector in the esophagus near the region of the tracheal bifurcation. Although the probe was developed specifically for counting plutonium, it can be readily applied to counting other isotopes emitting low-energy photons, such as ^{131}I, ^{125}I, ^{197}Hg, ^{111}In, ^{67}Ga, ^{133}Xe, ^{99m}Tc, etc.

The purpose of this paper is to describe both the physical features and the performance of a fiber-optic-coupled scintillation counter and to compare it with similar solid-state probes. Although fiber-optic probes have previously been built for dosimetry,[2] the present probe is designed for low-background, low-energy, high-sensitivity photon counting.

II. DESCRIPTION OF PROBE

Coupling a scintillation crystal to a photomultiplier with fiber optics causes a (80%–90%) loss in the scintillation intensity reaching the photomultiplier. Photons from the scintillations exit the crystal in nearly random directions and because of the small numerical aperture (limited acceptance cone) of the fiber optics only a small fraction of the photons are totally reflected from the sides of the fiber optics and conducted by the fibers.

Losses due to Fresnel reflection at the interfaces and losses in transmission also reduce the number of transmitted photons; however, these losses are insignificant with short fiber-optic bundles compared to the limited acceptance angle. Fiber optics with larger numerical apertures are available; however, their transmission around the 4000-Å peak of NaI(Tl) is unacceptably low. This loss of photons is serious at low energies because it reduces resolution and moves the scintillation spectra into the noise region of the phototube. For low-background counting and an improved signal-to-noise ratio, the background from the photomultiplier must be reduced. This was accomplished by selecting a low-noise high-gain phototube, magnetically limiting the active area of the photocathode, and by reducing the number of fast rise-time pulses by a pulse-shape discrimination technique.[3] Use of these techniques reduces the background count rate in the energy range from 6.4 to 40 keV by a factor of 50 or more (see Ref. 3).

A sectional drawing of the detector and probe is presented in Fig. 1. The NaI(Tl) crystal, surrounded by MgO, is encased in a thin aluminum shell (0.25 mm) with a glass optical window sealed in place by epoxy. The detector is coupled to the fiber-optic light guide by an optically transparent epoxy, an aluminum ferrule for mechanical strength, and an optically opaque epoxy which also occludes light and fills the remaining void.

The fiber-optic light guide is constructed of a 4.7-mm-diam bundle of 50-μm glass fibers having a numerical aperture of 0.66. The ends of the bundle are secured, bonded, and polished in stainless-steel ferrules to which an opaque outer sheath of polyvinyl chloride is attached.

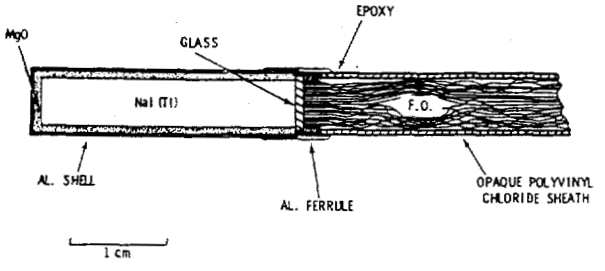

FIG. 1. Sectional drawing of probe.

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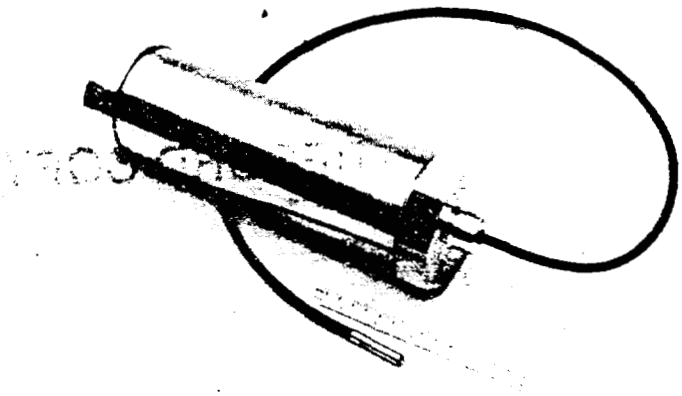

FIG. 2. Photograph of probe showing the detector, fiber optics, and photomultiplier housing. The sensitive volume is in the aluminum shaft shown next to the ruler.

The completed assembly is coated with a pure polyurethane resin and cured slowly at room temperature and high humidity. This material protects the probe from the caustic fluids within the digestive tract and softens the edges and corners of the probe. The probe cannot be autoclaved because of the thermal sensitivity of the NaI(Tl) crystal; however, gas sterilization has been used without degrading the performance or longevity of the probe.

Figure 2 is a photograph of a completed probe attached to a photomultiplier housing. An RCA 8850 photomultiplier with a 2-in. photocathode and the factory-recommended divider string are located within the housing. This unit uses a 2.5-cm-long by 4.7-mm (450 mm^3)-diam crystal coupled to the photomultiplier by 60 cm of fiber optics. Three other probes using a 90-cm-long fiber-optic coupling have been built and resulted in performance comparable to the present unit. Probes could be built in a large variety of sizes and shapes constrained primarily by light collection in the crystal geometry selected and by light transmission of the fiber optics.

III. PROBE PERFORMANCE

For photons of less than 80-keV energy, the calculated absorption in the detector for normal incidence is 100%; however, for low-energy photons attenuation in the aluminum encapsulation becomes important. For example, the intensity of the uranium L x rays emitted in the decay of plutonium (13.6, 17.0, and 20.2 keV) is reduced 30% by the 0.25-mm thickness of the aluminum encapsulation. On the other hand, at photon energies above 80 keV, the efficiency begins to fall and is reduced to 65% at 150 keV.

Figure 3 shows ^{99m}Tc spectra taken with the probe. The poor energy resolution is caused by light losses resulting from the fiber-optic coupling of the crystal as mentioned previously. Measured photon-energy resolution at 59.6 keV with the probe crystal directly coupled to the photomultiplier is 17%; however, with the fiber optics it is reduced to 54%.

Measured count rates (energy from 10 to 150 keV) for various sources located 5 cm from the probe in air are listed in Table I. The shielded and unshielded background count rates over three energy intervals are listed in Table II. Although pulse-shape discrimination does not appear to be important in reducing the background count rate for the probe, this is not generally the case. This was a phototube with excellent low-noise characteristics and tests on other phototubes required the use of pulse-shape discrimination for ultimate background count-rate reduction.

The count rate with the scintillation probe immersed in a 500-ml beaker containing a solution of ^{99m}Tc (Fig. 3) is 5.6×10^6 cpm/μCi/ml. The count rate for a GaAs biomedical probe under similar conditions is 200 cpm/μCi/ml,[4] and results published for a large-volume CdTe biomedical probe[5] (50 mm^3) indicate a count rate of 6.3×10^4 cpm/μCi/ml in a more favorable geometry (smaller beaker). It must be noted that both the CdTe (50 mm^3) and GaAs (0.19 mm^3) probes have much smaller volumes than the scintillation probe with its active volume of 450 mm^3.

Normalizing the above count rates to the same volume

TABLE I. Count rate obtained with various sources in a standard geometry.[c]

Source	Count rate (10–150 keV) (cpm/μCi)	Principal energies (keV)
Am 241	3870±28	17, 60
Co 57	5330±56	122, 136, 14.4
Cd 109	5670±46	23, 88
"Mock" I 131	8440±41	30, 80
Pu 239[a]	70± 2	17
Pu 239[b]	462± 5	17, 60

[a] Isotopically pure Pu 239.
[b] Typically encountered Pu-239 with other Pu isotopes and Am 241 daughter.
[c] Point sources 5 cm from probe in air.

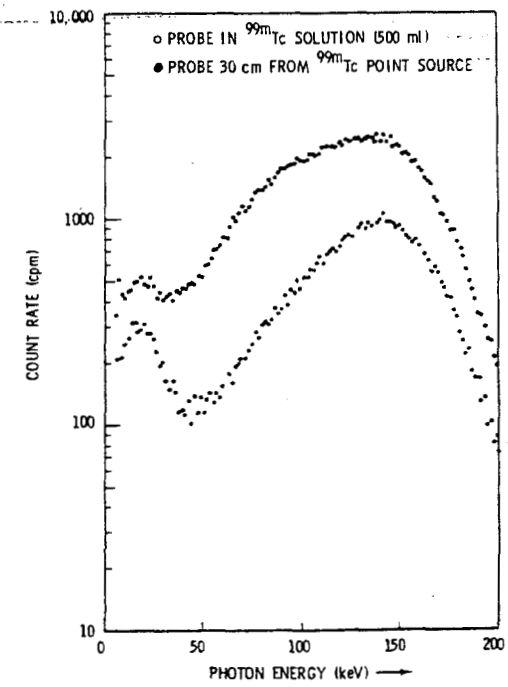

FIG. 3. Spectra taken with sources of ^{99m}Tc. (The reduced energy resolution of the system is apparent and the effect of scattering in a low-Z medium can be seen.)

TABLE II. Background count rates in various energy intervals with and without shielding and/or pulse-shape discrimination.

Pulse-shape discrimination	Shield[a]	Background count rates (cpm) 10–40 keV	40–90 keV	90–150 keV
No	No	19.9±1.2	51.9±1.9	73.9±2.2
Yes	No	15 ±1	56.0±1.9	70.7±2.2
No	Yes	7.1±0.7	1.3±0.3	7.5±0.7
Yes	Yes	3.9±0.5	1.5±0.3	9.1±0.8

[a] 3.9-cm lead shield around detector.

still shows an order of magnitude greater sensitivity for the scintillation probe. Encapsulation, energy-cutoff point, charge-collection efficiency, geometry, and detector Z will influence the comparison of probes in such a geometry in addition to detector volume.

Previous esophageal probes for plutonium detection were built using avalanche silicon diodes as detectors.[6] These diodes make excellent detectors, but are limited in ultimate sensitivity by their low absorption of incident photons. The calculated intrinsic efficiency of these diodes is approximately 12% based on normally incident L x rays from the decay of plutonium. The diodes have extremely low background count rates (~1 cpm), which help compensate for their poor efficiency. In contrast, thin detectors of NaI(Tl), with approximately 100% efficiency, have long been used to detect these x rays.

Table III shows typical background count rates, sensitivities, and minimum detectable amounts (MDA) for avalanche probes and the 450-mm³ NaI(Tl) probe in a thorax phantom representing the human tracheobronchial lymph node geometry. To visualize the counting geometry, consider three small masses located in the angles of the tracheal bifurcation. If these three masses (sources), located about 2 cm apart, are considered to be at the vertices of the base of a tetrahedron, then the center of the detector would be in the position of the remaining vertex, but within the esophagus. The three masses of lymph nodes weigh a few grams with the largest mass in the angle formed by the two major bronchii. The phantom simulates this geometry with sources in place of the lymph nodes. The avalanche diode probes in Table III consist of a three diode array used to improve angular response,[6] and they have a total sensitive volume of about 15 mm³. Again, the larger volume of the NaI(Tl) probe largely accounts for the improved sensitivity; however, one must realize large volumes of suitably configured solid-state detector materials are not feasible at this time. We are comparing the best solid-state probe with our scintillation probe based on this specific application.

TABLE III. Background, sensitivity, and minimum detectable activities for intraesophageal plutonium detectors.

Probe	Detector volume (mm³)	Background (B) count rate (cpm)	Sensitivity (S) (cpm/nCi)	Minimum detectable amount[a] (nCi)
Avalanche 1	15	5.05	0.025	52
Avalanche 2	15	5.56	0.039	34
Fiber optics:	450			
x-ray energies (17 keV)		18.4 (9.20)[b]	0.807	3.0
γ-ray energies (60 keV)		38.7 (3.06)[b]	1.22	2.9

[a] Calculated at 90% confidence level with a 20-min counting period; i.e., $MDA = (2.56/S)(1/20B)^{\frac{1}{2}}$.

[b] Values in parenthesis are for detector shielded by 3.9 cm of lead.

Plutonium, as usually encountered, consists of its various isotopes in certain proportions and ^{241}Am,[7] a daughter of ^{241}Pu. The 59.6-keV γ rays from ^{241}Am can reach the esophageal detector from the lung, which will usually necessitate a correction of the count rate with the scintillation probe, whereas the L x rays from the lung are heavily attenuated. Avalanche probes have a poor efficiency at 59.6 keV (~½%), which reduces the interference, while pulse-height selection can be used to control the interference for the scintillation probe. The sensitivities at 59.6 keV listed in Table III are for materials located in the lymph nodes.

IV. PROBE APPLICATION

The 450-mm³ scintillation probe has been used to count plutonium translocated to the tracheobronchial lymph nodes of dogs which had been exposed to a respirable aerosol of $^{239}PuO_2$.[1] Previous results[5] with the avalanche probes indicated that a sensitivity of greater than 1 cpm/nCi could be expected based on the relative sensitivities of the two types of probes. Figure 4 shows a scan of the count rate versus position referenced to the incisors. The relatively sharp peak in count rate would not exist if material in the lymph nodes were not being counted. If material were not in the lymph nodes, a constant but slight elevation in count rate would be evident from material in the lungs. Radiographs were used to verify that the maximum count rate corresponded to the lymph node region. The lymph node geometry is similar to that depicted for the humans; however, the distance between the three tracheobronchial lymph nodes is reduced to 1.5 cm and the total mass of the nodes is less than ½ g. Four dogs were counted and, based on thoracic lymph node burdens estimated by extrapolation from measured lymph node buildup in sacrificed animals and whole-body counts, the sensitivity varied from 0.45 to 1.9 cpm/nCi with an average of 1.2 cpm/nCi. These results appear reasonable considering the potential for large errors in the extrapolations because of biological variability and possible errors in whole-body counting. Actual sensitivities can be expected to be somewhat better since tracheobronchial burdens constitute only a portion of the thoracic (tracheobronchial, mediastinal, sternal) lymph node burdens used in estimating sensitivities.

Two probes were built for counting plutonium in the surface tissues. These probes have an end window (3 mm diam) and a sensitive volume of 25.3 mm³. The probes were about 20 times more sensitive than comparable sized solid-state avalanche probes. Although these probes were not intended for *in vivo* applications, they were used to study the feasibility of using a fiber-optic coupled scintillator in conjunction