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Evaluation of Thin CaF_2 (Eu) Scintillator for Detecting Tritium

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EVALUATION OF THIN CaF_2 (Eu) SCINTILLATOR FOR DETECTING TRITIUM

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ABSTRACT

The primary objective of this project was to investigate the feasibility of using a CaF_2(Eu) scintillator for detecting low-energy beta particles from tritium. A proof-of-principle detector was designed for flowing tritium-spiked nitrogen gas across the surface of a thin scintillator, which was optically coupled between two low-noise photomultiplier tubes. Electronics for operating the two photomultiplier tubes in coincidence eliminated most of the tube noise pulses and allowed detection of the small pulses from the low-energy tritium beta particles.

1. INTRODUCTION

The primary objective of this project was to investigate the feasibility of using a previously suggested CaF_2(Eu) scintillator for detecting tritium (^3H)[1]. In order to evaluate the usefulness of CaF_2(Eu) for this application, it was necessary to first determine its counting sensitivity for low-energy beta radiation from tritium. The maximum energy beta particles from tritium is 18 keV, and the average energy is about 6 keV, making it very difficult to discern small electronic pulses from these low-energy particles in the presence of the noise pulses generated in the photomultiplier tubes. This situation is improved by using a coincidence circuit to generate a gate pulse only when a pulse from both tubes occurs simultaneously, hence rejecting most of the tube noise pulses. After determining the counting sensitivity, a detector of adequate size to detect 10^{-13} Ci/mL in air (which is the goal set by the Navy's RADIAC program) can be designed.

In order to simplify the initial proof-of-principle detector and test equipment setup, a beta source was prepared by depositing a small amount of ^{63}Ni on a flat aluminum plate. After the source was dried, a thin (0.00015 in.) mylar film was placed over the plate and glued around the edges to contain the radioactive material. This was a much more convenient source to handle during preliminary tests than tritium, which is in a gaseous state at room temperature. The preliminary data from the ^{63}Ni source (maximum beta energy of 66 keV and average energy of 17 keV) indicated that the lower-energy beta particles from tritium can be detected using this technique.

2. DESCRIPTION OF THE SCINTILLATOR

The optimum thickness of CaF_2(Eu) to absorb the maximum energy beta particles from tritium and yet not absorb enough energy from background gamma photons to generate a detectable pulse is approximately 920 $\mu g/cm^2$ (ref. 2). A scintillator was prepared by evaporating CaF_2(Eu) onto both flat surfaces of a 2-mm-thick pyrex glass plate. The layer of scintillator deposited on the glass plate had an area density of 900 μg/cm^2 on one side and 1000 μg/cm^2 on the other side, which is near the desired optimum thickness.

One conceptual design[1] for a tritium monitor consists of several glass plates (light guides) coated with CaF_2(Eu) arranged in parallel with approximately 0.25-in. spacing with two low-noise and high-gain photomultiplier (PM) tubes optically coupled to opposite edges of the glass plates as shown in Fig. 1. In order to investigate this concept, a glass plate was coated on one side with CaF_2(Eu). This plate was used to determine the fraction of light lost when the tubes are coupled to the edges of the light guides compared to the light when coupled to the flat side. Pulse amplitude measurements were made both while the glass plate was optically coupled directly on the flat face of one PM tube and while one edge of the plate was coupled to the PM tube. The ratio of the average pulse amplitudes with the scintillator flat on the face of the tube and exposed to the ^{63}Ni source compared with the average pulse amplitude with the edge of the scintillator and glass plate coupled to the tube was 2.6. Pulse amplitude spectra for both arrangements are shown in Fig. 2. This measurement indicates that a significant portion of light is lost when tubes are coupled to the edge of the light guide; but by using high-gain, low-noise PM tubes and coincidence electronics (which will be explained later), sufficient signal is still available to detect the low-energy beta particles when tubes are coupled to the edges. This arrangement allows a large area of scintillator to be coupled to fairly small PM tubes, whereas if the scintillator was coupled flat on the tube, a very large PM tube would be required.

3. DESCRIPTION OF COINCIDENCE ELECTRONICS

The inherent dark-noise pulses from a photomultiplier tube are about the same amplitude as some of the smallest pulses generated in CaF_2(Eu) by low-energy beta particles. The majority of these randomly occurring, undesirable noise pulses can be eliminated by using two photomultiplier tubes to view the same scintillator and operating them in coincidence mode, whereby only a simultaneous pulse from both tubes will be recorded. A block diagram of the system is shown in Fig. 3. The fraction of total events counted in the coincidence operation depends on the resolving time allowed in the coincidence unit for the two pulses to coincide and supply a gate pulse to the multichannel analyzer. The resolving time of 2.0 μs gave best efficiency in these measurements.

4. TRITIUM TESTS

A different scintillator assembly was designed to allow nitrogen gas spiked with tritium (tritiated methane) to flow through the scintillator cell as beta particles interact with the CaF_2(Eu). Figure 4 illustrates the design. A photograph of a pulse-height spectrum from tritium-spiked gas is shown in Fig. 5. The two parameters to be considered in estimating the sensitive volume of the flow cell are range of the beta particles and area of the scintillator. The area of the scintillator used is 18.6 cm^2, but the range varies with the energy of the beta particles (discussed in the following section). Beta activity of the ^3H-spiked nitrogen gas tested was 2.6 nCi/mL, and with optimum electronic adjustments, the count rate in coincidence operation was about 21 counts/s.

5. RATIONALE FOR ESTIMATING THE BETA ACTIVITY ON THE SCINTILLATOR

In order to calculate an approximate beta exposure to the scintillator from the tritium-spiked gas, the beta energy distribution from tritium and the range of the beta particles were investigated. A tritium spectrum shown in Fig. 6 and reported by Curran, Angus, and Cockroft[3] was selected as representative of the beta energy distribution. This distribution was divided into three energy brackets centered on values of energies for which published range data are available. The three energy brackets selected are 5 ± 3 keV, 10 ± 2 keV, and 15 ± 3 keV, covering an energy span of betas from 2 to 18 keV. An estimated 20% of the betas have energy <2 keV and after light losses will not generate a sufficient pulse in each PM tube to be counted in the coincidence mode. Therefore, detectable beta activity >2 keV energy is estimated to be 80% of the total.

A population or percentage of beta activity in each energy bracket was estimated from the spectrum curve to be as follows: approximately 48% of the total beta population is in the energy bracket of 2 to 8 keV, about 20% is in the bracket of 8 to 12 keV, and about 12% is in the remaining bracket of 12 to 18 keV. These values along with the range of the median energy beta particles of each bracket are listed in Table 1.

By summing the products of the area of the scintillator (18.6 cm^2), the depth of penetration and the beta activity in each bracket, an estimated number of betas striking the scintillator can be calculated for each bracket: 8, 11.5, and 14, respectively.

Therefore, total betas estimated to reach the scintillator $\approx 8 + 11.5 + 14 \approx 33.5$ betas/s.

The integral counts from flowing the tritium-spiked nitrogen gas through the experimental detector was 21000 counts/1000 s. Hence, the detector counted approximately 62.7% of the betas that were estimated to contact the scintillator.

Table 1. Beta activity in energy brackets

	Energy brackets (keV) 2-8	8-12	12-18
Median energy (keV)	5.0	10.0	15.0
Median range (csda)* in nitrogen (mg/cm^2)	0.085[4]	0.285[2]	0.585[2]
Median projected range (~1/2 median range)[5]	0.043	0.142	0.292
Penetration depth in nitrogen (cm) (median projected range/density at 23°C)	0.037	0.124	0.254
Fraction of total beta activity in bracket	0.48	0.20	0.12
Beta activity in bracket dis/s/mL (tritium concentration = 2.6 nCi/mL)	46.0	19.0	11.5
Only 50% of betas will penetrate the thickness of sample listed above β/s/mL (by definition of median range)	23.0	9.5	6.0
Geometry fraction--only 50% of betas migrate toward scintillator, therefore, specific activity in direction of scintillator = β/s/mL	11.5	5.0	3.0

*csda: Continuous slowing-down approximation.

6. CONCLUSION

The CaF_2(Eu) scintillator in conjunction with the coincidence technique used in this investigation is a very stable and simple method for detecting low-energy beta radiation. In this feasibility experiment, most of the beta particles contacting the scintillator were detected, thus it is obvious that the only parameter that can be increased to improve the sensitivity is the area of the scintillator. In order to detect low activity of 10^{-13} Ci/mL in air with 10% accuracy within 2 min., a very large area (2 m^2) detector will be required.

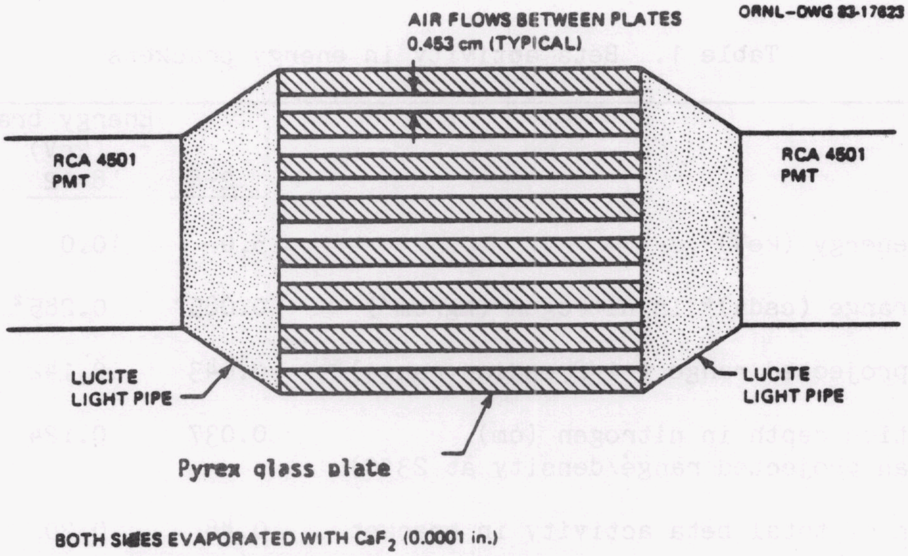

Fig. 1. Conceptual design for several glass plates coated with scintillator optically coupled between two photomultiplier tubes.

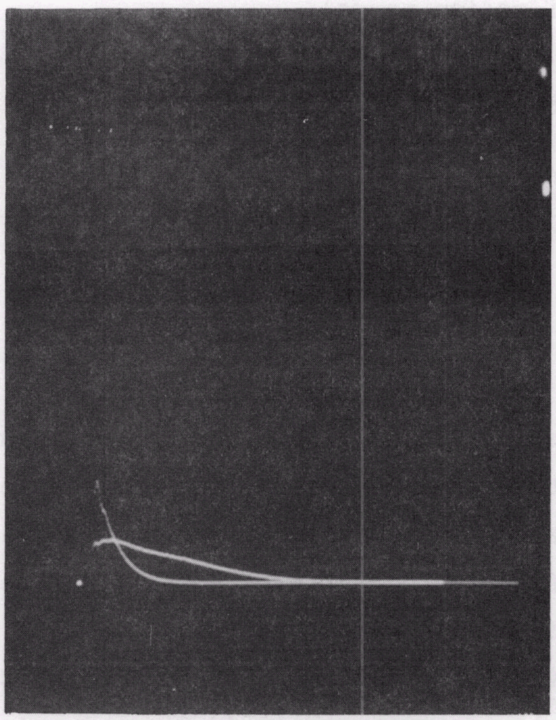

Fig. 2. Pulse amplitude spectra with scintillator flat on the face of the tube (upper-right curve) compared with the spectra with the edge of the scintillator coupled to the face of the tube (lower curve.)

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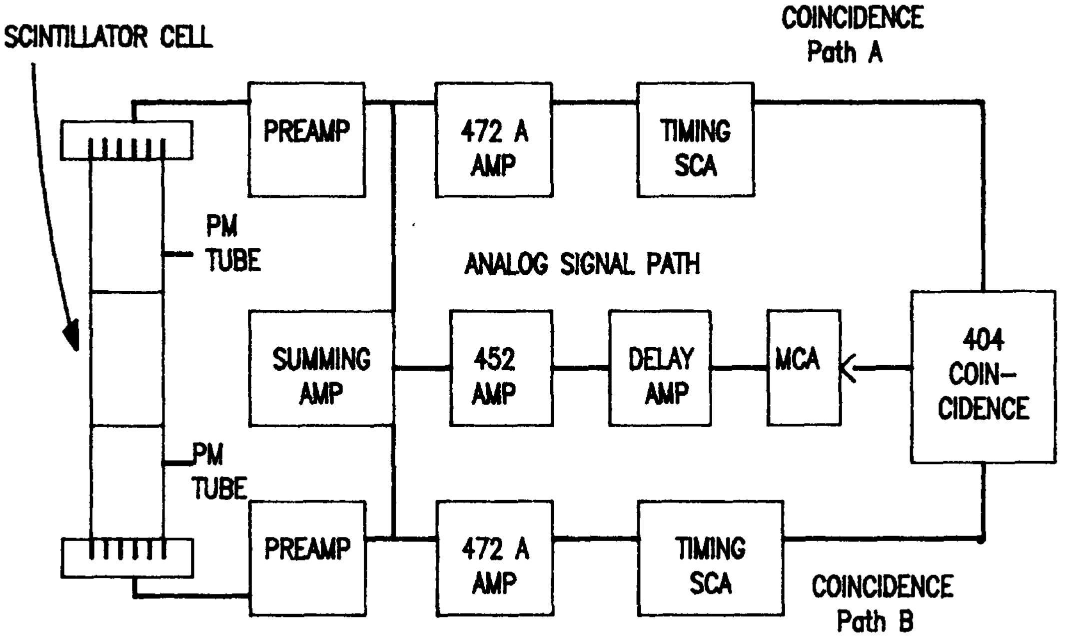

Fig. 3. Block diagram of the electronics for a detector assembly using two photomultiplier tubes. The tubes are coupled to opposite edges of the scintillator and operate in coincidence mode to prevent pulses occuring outside the scintillator from being counted.

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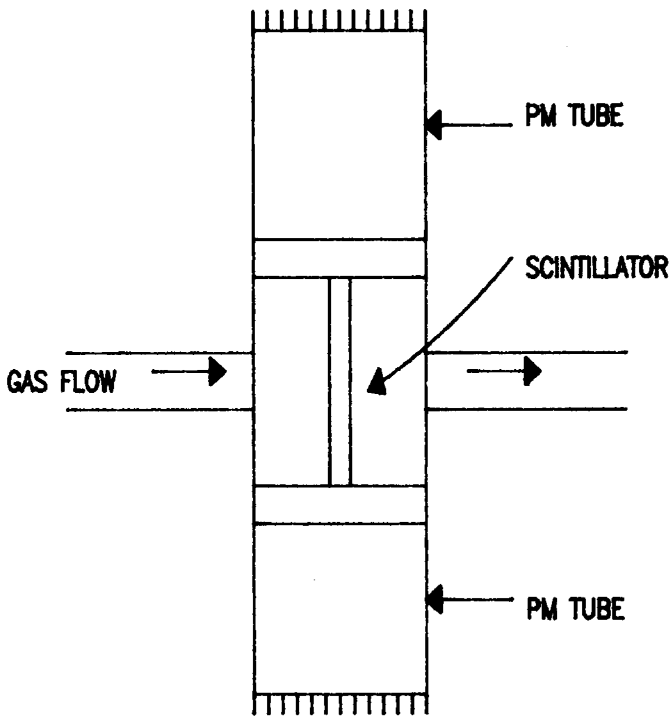

Fig. 4. Gas flow cell for testing ^3H sensitivity. The pyrex plate is coated with CaF_2(Eu) on both sides and in the center of the cell; the edges are coupled to two photomultiplier tubes.

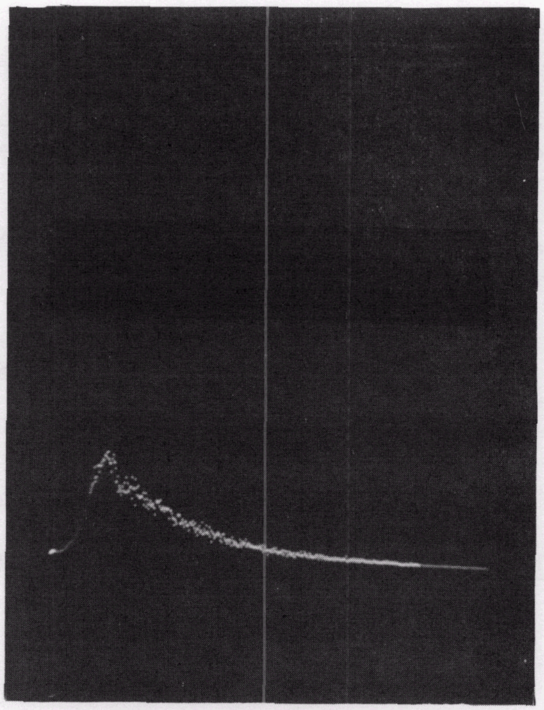

Fig. 5. Pulse-height spectrum from the tritium-spiked gas.

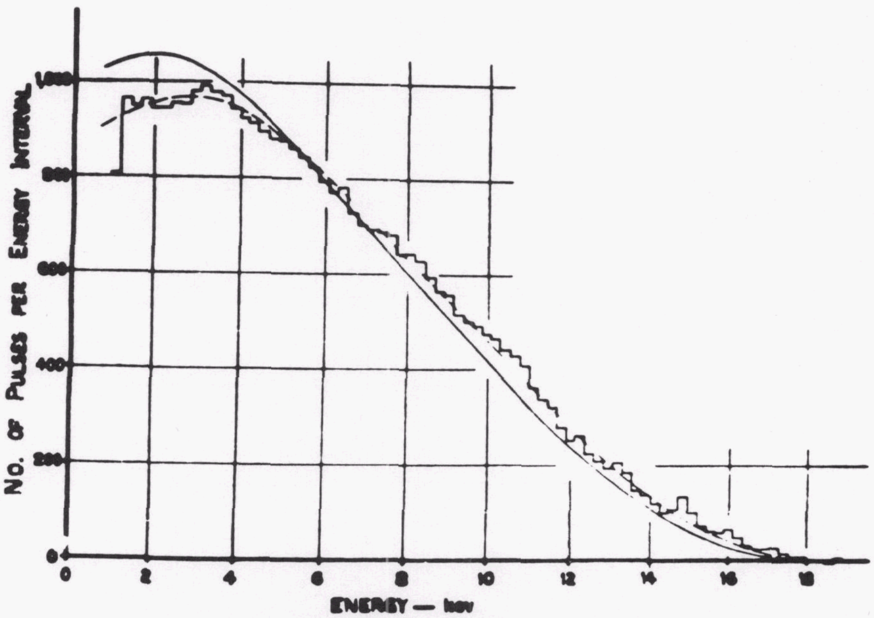

Fig. 6. Beta energy spectrum of tritium. Source: S. C. Curran, J. Angus, and A. L. Cockroft, "Investigation of Soft Radiations-II: The Beta Spectrum of Tritium," Department of Natural Philosophy, University of Glasgow, Philosophical Magazine 40, 1949.

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