A BETA-RAY MICROSCOPE

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1. Introduction

The potential value of radioactive tracers in many fields of physical, chemical and biological investigation was recognized very soon after the discovery of natural radioactivity. Not, however, until the nuclear reactor made the production of many artificially radioactive isotopes possible on a large scale did the use of radioactive tracers reach anything approaching its present state of development.

The great power of this method of investigation lies in its potential ability to tag a particular atom and thus allow the identification of this atom as an individual.

Thus, for example, the mobility of individual atoms or groups of atoms may be studied in a dynamic system by introducing radioactive atoms into some part of the system and making measurements which determine their rate of appearance at other points in the system.[1] Studies of the arrangement of atoms in relatively static systems may also be made by incorporating a small portion of a radioactive isotope of the atom of interest into the system and making measurements to determine the structural arrangement of the radioactive atoms.

That studies made with the radioactive isotopes of an element give information applicable to stable isotopes of this element is due to the fact that in many chemical and physical situations the radioactive isotopes of an element behave in almost the same way as the stable isotopes.

The main determinant in the success and accuracy of any method of analysis which uses radioactive tracers is the precision with which measurements that indicate the presence and concentration of the radioactive isotopes may be made. In some cases only an indication of the concentration of the radioactive isotopes in a rather large region may be sufficient. This is true, for example, in some medical applications such as the study of iodine uptake of the thyroid. In other cases such as the study of the microstructure of metals, it is desirable to be able to locate the sources of radiation to within a few microns.

This paper is concerned with the study of a method for

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attaining medium precision (of the order of 100 microns) in the location of radioactive isotopes in relatively static systems. The method proposed makes use of a thin transparent scintillator or phosphor as the primary detector of the beta particles emitted by radioactive isotopes. Fig. 1 shows the phosphor in contact with the specimen being examined. Some of the energy of the high energy beta particles from the specimen is converted within the phosphor into visible radiation. The light from an area of the scintillator selected by an optical system is focused on a suitable detector. The location and the amount of the light from the selected area of the thin phosphor layer indicates the presence and concentration of radioactive atoms in that part of the specimen directly below.

An analysis of this method has been made, and an attempt has been made to compare the analysis with experiment. With the method that has resulted from this study it is possible to locate certain radioactive sources to within at least a 100 micron diameter field on or near the surface of a thin flat section of a specimen containing radioactive atoms. Such a determination is made in a matter of a few minutes, since the method involves determination of the instantaneous rate of decay of isotopes in the field.

2. Expected Resolution and Sensitivity of Proposed System

Because of the high specific ionization caused by alpha particles in their passage through matter, the best resolution in the location of radioactive isotopes can be attained for those isotopes that emit alpha particles. Since the alpha emitting isotopes are restricted to a relatively few elements it is desirable that a system which is to be generally useful as an investigative tool be capable of detecting other particles.

The nuclear reactor has made many beta emitting isotopes readily available to investigators. For a known energy the maximum range of a beta particle can be fairly accurately predicted.[2] Since beta emitting isotopes lend themselves reasonably well to precise location problems, it was decided to design a system having maximum efficiency in the detection of betas.

Suppose one wishes to determine the pattern of the distribution of the radioactive atoms in the surface structure of a specimen. Since the emitted betas are high speed charged particles, some method of focusing these particles upon a suitable detecting device in order to produce an image might seem to be a possibility. However, except for those few isotopes in which conversion electrons are produced, the electrons which are emitted may have any energy from zero up to a maximum value typical of the particular isotope considered. Since the emitted betas have a wide continuous spectrum of energies, it is almost impossible to focus them electromagnetically in order to obtain image

formation.[3]

The complication in image formation introduced by the wide range of beta energies is avoided by the use of a scintillator as the primary detector of the high energy particles as illustrated in Fig. 1. The image formation is now considerably simplified, since it is in terms of light with a narrow range of energies rather than in terms of the betas themselves. Since the amount of light produced per particle-phosphor interaction is too small to be detected by the unaided eye, an optical lens system selects the light coming from a specified area of the scintillating layer and focuses it on the photocathode of an electron multiplier type phototube. The phosphor-phototube combination with suitable electronic circuitry serves as a radiation detector.

In order to achieve maximum resolution with the proposed method of locating radioactivity it is important that both source and detecting layer be thin.[4] The phenomenon which is actually measured is the path or paths of the betas. Resolution thus depends upon how sensitive the method of measurement is to a variation in the intensity of paths within the scintillator as a function of distance from a source. Since the greatest variation in radiation intensity with distance from a point source occurs nearest the source, it is important for high resolution that all possible sources be as near the scintillator as is practicable. This implies both sources and scintillators as thin as can be prepared without changing their normal physical characteristics.

The phosphor indicates the passage of a beta particle by absorbing the particle energy and converting some of it into electromagnetic radiation in the visible or near visible region. In order that enough light reach the phototube through the collector it is important that the conversion efficiency of the phosphor be high. In a system which achieves a resolution of 100 microns the phosphor can not be much thicker than this. The more efficient organic phosphors will not absorb all the energy of even a maximum energy Carbon 14 beta in this thickness. (The Carbon 14 beta with maximum energy of 155 kev has a range in anthracene of about 240 microns.) It is clear that a compromise has to be reached between sensitivity and resolution for all but the very low energy betas.

The sensitivity and resolution might both be improved if a very dense phosphor were available. Cadmium tungstate was tested for this reason; but was found to give results much inferior to those given by anthracene. Of all the organic phosphors tested anthracene proved to be the most sensitive in thin sections.

In order to establish the resolution it is necessary to

know how close to the particle path the light is produced. The passage of a high energy particle through anthracene and the energy conversion process producing the scintillation radiation is apparently a property of the molecular structure. Anthracene in common with most other organic phosphors is characterized by conjugated double bonded carbon atoms in benzene rings. The passage of the particle probably disrupts the arrangement of the electrons in the closed ring molecules, perhaps by producing changes in the electron spins and the energy distributions associated with the alternating single and double bonds.[5] The light is thus probably emitted by the disturbed molecule, very near the path (certainly within 1 micron) of the beta particle. The light source resulting will be a line source with intensity that is in many cases almost uniform along the path length.

A microscope objective lens placed with its axis perpindicular to the phosphor surface selects a circular field on the phosphor surface. The size of the selected field can be controlled by placing a circular aperture at the image plane of the objective. The amount of light collected from a light source within the selected field depends both upon the numerical aperture of the objective lens and the index of refraction of the phosphor. An analysis based upon geometric optics, and assuming a flat polished phosphor surface, shows that the maximum fraction of the light produced at a source point within the phosphor is transmitted through the optical system if the source point is on the objective axis. Only a negligible fraction of the light produced outside a cylindrical section determined by the real field of the optical system at the upper surface of the phosphor (see Fig. 1) is transmitted by the optical system. Hence the resolution can be determined by the size of the field-determining diaphragm placed at the objective image plane.

The conversion efficiency for anthracene was computed from data given by Hofstatder, Kallman, and Gillette to be about 7.5 per cent for betas.[6,7,8] This figure is what Kallman calls the "physical efficiency" of the phosphor, that is, the actual efficiency with which the beta energy is converted into scintillation radiation. A 97x American Optical microscope objective with a numerical aperture of 0.828 was used as a light collector. Calculations were made assuming all the visible radiation to be emitted at a point on the objective axis. Mean photon energy was taken as that of a photon at the radiation peak of anthracene at 4440 A.[9] Under the optimum condition of a maximum energy beta with all its energy absorbed in anthracene, results indicating the number of photons delivered to a phototube cathode may be obtained.

Table 1 gives the results for an anthracene scintillator and three different isotopes: Cobalt 60 with a maximum beta energy of 310 kev, Carbon 14 with a maximum of 155 kev, and Nickel 63 with a maximum of 60.7 kev.

Table 1. Mean Photon Yield Per Event

Isotope	Photons to Single Photocathode	Photons to Each of Two P.cathodes	Electrons from P.cathode
Co 60	393	180	17.85
C 14	197	90	8.93
Ni 63	77	35	3.47

For reasons to be discussed in Section 3 a light splitter was used to divide the light from the objective into two approximately equal parts. As discussed in Section 3 the transmission of this light splitter was about 92 per cent. The third column of Table 1 gives the resultant number of photons to each of two phototubes when the light splitter is used.

Calculations using the manufacturer's data indicate that the quantum efficiency for the photocathode of the type 1P21 phototube at 4440 A is about 9.92 per cent. Column four of Table 1 gives the average number of electrons from the photocathode for an interaction involving a peak energy beta. On the average, of course, an event might result in a considerably smaller number of photons than that indicated in this table since the mean beta energy is roughly one third the maximum.

Since the assumptions of a point source and maximum beta energy made in getting the results listed in Table 1 are not usually met in practice, these results serve only as an indication of an optimum pulse size to be expected in practice. Thus the results of column four would seem to indicate that enough light should reach each of two phototubes from interactions of some of the betas from Cobalt 60 and Carbon 14 to give a detectable signal. Nickel 63 would seem from these results to be near the borderline of detection. Experiment shows that it is indeed possible to detect both Cobalt 60 and Carbon 14 within a 75 micron diameter field on the surface of a section of anthracene 50 microns thick. Nickel betas can also be detected, but the detection efficiency for Nickel is low.

3. The Noise Problem

As is well known to anyone who has had occasion to use electron multiplier type phototubes, many pulses appear at the anode that are not caused by the release of photoelectrons at the cathode. If a signal is represented by the release of photoelectrons from the cathode, some method must be found for distinguishing between these signal pulses and the spurious pulses which are frequently referred to as noise.

Since many of these noise pulses result from thermal emission of electrons from the photocathode and from the dynodes,

(Morton has reported 9500 electrons per second thermally emitted from the photocathode at room temperature as being typical of a 1P21 phototube.[10]) cooling of the tube is a satisfactory laboratory method of reducing noise. Cooling, however, introduces some rather formidable problems in insulation because of the condensation of moisture on the cooled parts. Since it was felt desirable to eliminate these problems in this equipment, a different approach, that of using two phototubes in coincidence, was tried.

The use of two phototubes in coincidence to reduce the effect of the random noise pulses of an individual tube is a well developed technique in the field of scintillation counting.[11,12,13] In most cases the two tubes are arranged so that each tube views the phosphor directly. A part of the light generated in the phosphor falls on each tube individually. The unique feature of the method of detection used in the microscope described here is that the optical system gathers the light from a small selected area of the phosphor, and focuses it on a light splitter. The light splitter divides the light approximately in half and each half goes to a phototube.

Figure 2 is a simplified diagram of the optical system and the light splitter. Note that for a point slightly off the objective axis the light split will not be exactly 50-50. The closer the source is to the objective axis the more nearly will the light going to the two phototubes be equal.

The light splitter used in the optical system is a right angle prism with the vertex of the right angle ground to a sharp edge. Aluminum was evaporated in a vacuum on the sides adjacent to the right angle in order to form a reflecting surface. The coefficient of reflection of aluminum evaporated on glass is about 0.92 at the radiation peak of anthracene.[14] Thus approximately 0.46 of the light from the objective is delivered to the phototube.

The use of a fairly conventional coincidence circuit having a resolving time of 0.25 microseconds with the system as presently designed decreases the background from several thousand counts a minute typical of one tube, to a few counts a minute for the two tubes in coincidence.

4. Description of Apparatus

A block diagram of the Beta-Ray Microscope is shown in Figure 3. The components will be described below.

1. The source is in the form of a thin flat section of a material containing the isotopes to be detected.
2. The phosphor is a thin section of an anthracene crystal placed in contact with the surface of the specimen being studied. Small crystals of from about 15 to 100 microns in thickness have been grown by evaporation from a large crystal and recondensation. Small sections about 200 microns thick and thicker

can be shaved from a larger crystal with a sharp razor blade.
3. The optical system gathers the light from a selected area of the phosphor, splits it approximately in half, and focuses it on the photocathodes of the phototubes. The light gathering lens is a standard 97x microscope objective. The optical system is supported by a modified microscope stand. The source and phosphor are positioned under the objective by a movable microscope stage. In place of the light splitter a standard ocular can be substituted thus allowing the specimen surface to be visually examined through the thin transparent phosphor.
4. The phototubes convert the light pulses striking their photocathodes into electrical pulses. Since the light being detected is of a very low intensity, they must be operated in complete darkness. In the present model of the microscope the source, the phosphor, the optical system, and the phototubes all are housed in a light tight enclosure. This enclosure may be removed for adjustments and for visual viewing of the specimen surface.
5. The preamplifiers are used here as impedance matching devices which allow the impedance across the phototube anode to be relatively independent of the linear amplifier input impedance.
6. The linear amplifiers amplify the pulses from the preamplifiers, approximately preserving the maximum amplitudes, but not necessarily the pulse shapes. This amplification is necessary in order that the pulses be large enough to properly operate the coincidence circuit.
7. The pulse amplitude selectors give no output for input pulses smaller than some selected amplitude. Each input pulse larger than this selected amplitude produces an output pulse of a uniform width and amplitude. This uniformity gives much more reliable operation of the coincidence circuit than do non-uniform pulses. The amplitude selectors can be set to eliminate many small noise pulses without appreciably affecting the signal pulses.
8. The coincidence circuit is used to reduce the noise background of the phototube detector as discussed in Section 3.
9. The recorder may count pulses over a selected time interval or it may be in the form of a count rate meter. Usually for weak sources the individual pulse count is preferable.
10. The phototube power supplies are stabilized high voltage supplies adjustable over a range of from 600 to 1050 volts. Individual supplies allow for differences in the two phototubes.

5. Results

Satisfactory detection of betas from Cobalt 60, Thallium 204, Carbon 14, and Nickel 63 has been achieved with anthracene crystals 50 microns thick. The selected phosphor field was 75 microns in diameter. Activities were of the order of 1 microcurie per sq. cm. Light collection was accomplished with a standard 97x microscope objective focused on the phosphor surface. The field was

defined by means of a circular aperture placed in the image plane of the objective.

In the case of Carbon 14 betas detection efficiencies as high as about 50 per cent have been achieved with a 200 micron thick anthracene crystal. The best detection efficiency achieved with Nickel 63 to date has been about 2 or 3 per cent.

Figure 4 is a plot of measured count rate against distance from the edge of a source. The source was Thallium 204 plated on nickel-plated copper. Source intensity was about 1.5 microcuries per sq. cm. The source was flat, about one cm. square. The zero point on the abscissa was the approximate edge of the metal as observed visually with the microscope objective and an ocular.

An improved model of the microscope is now under construction which will allow visual viewing of the selected field to be more easily accomplished. The adjustment of selected field diameter will also be easier on this model.

Additional investigation of plastic phosphors is also planned, because of the ease with which thin sheets can be fabricated. Those presently available have only about 40 per cent the conversion efficiency of anthracene, but it is possible that this can be improved.

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Figure Captions

Figure 1. Source Scintillator Arrangement

Figure 2. Simplified Diagram of Path of Light Through Optical System

Figure 3. Block Diagram of Apparatus

Figure 4. Measured Activity vs. Distance from Edge of Source. Source, Thallium 204, Plated on Nickel-Plated Copper. Anthracene Crystal 250 Microns Thick. 97x Objective. Real Field 75 Microns.

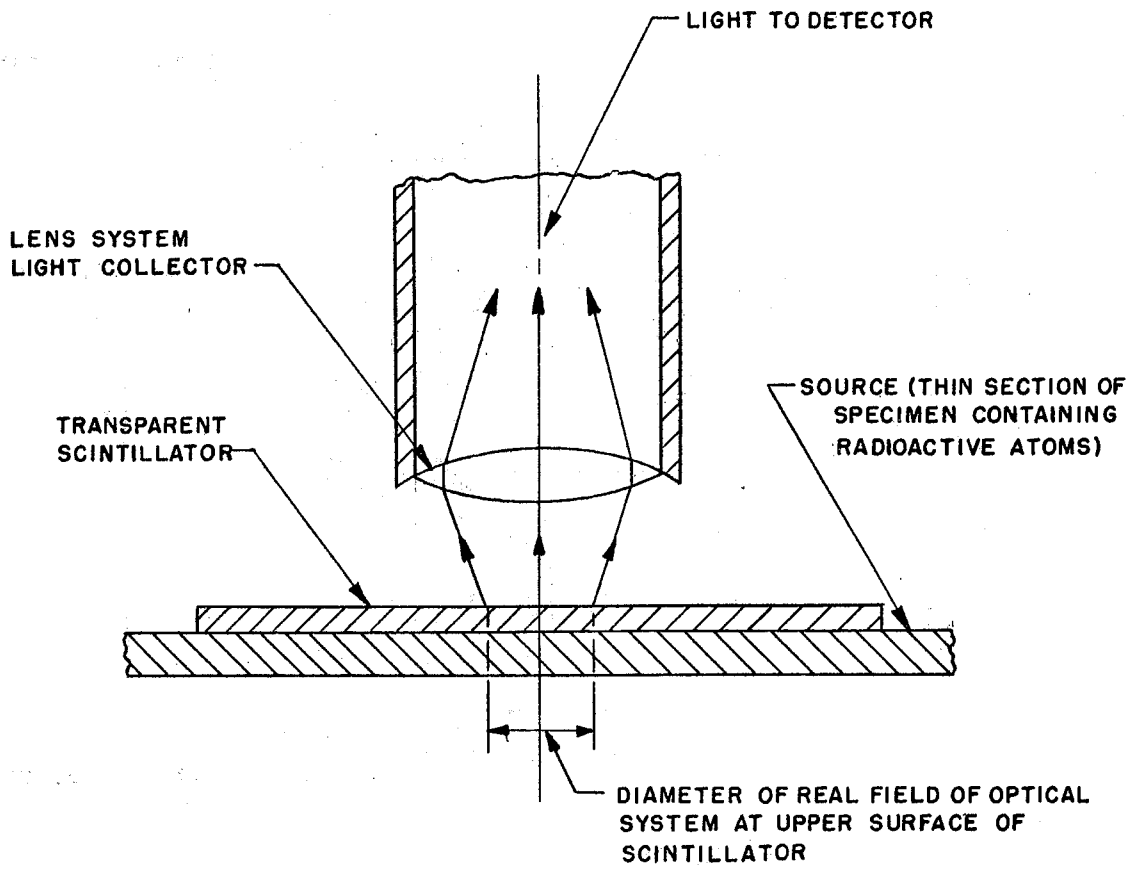

FIG. I

SOURCE SCINTILLATOR ARRANGEMENT

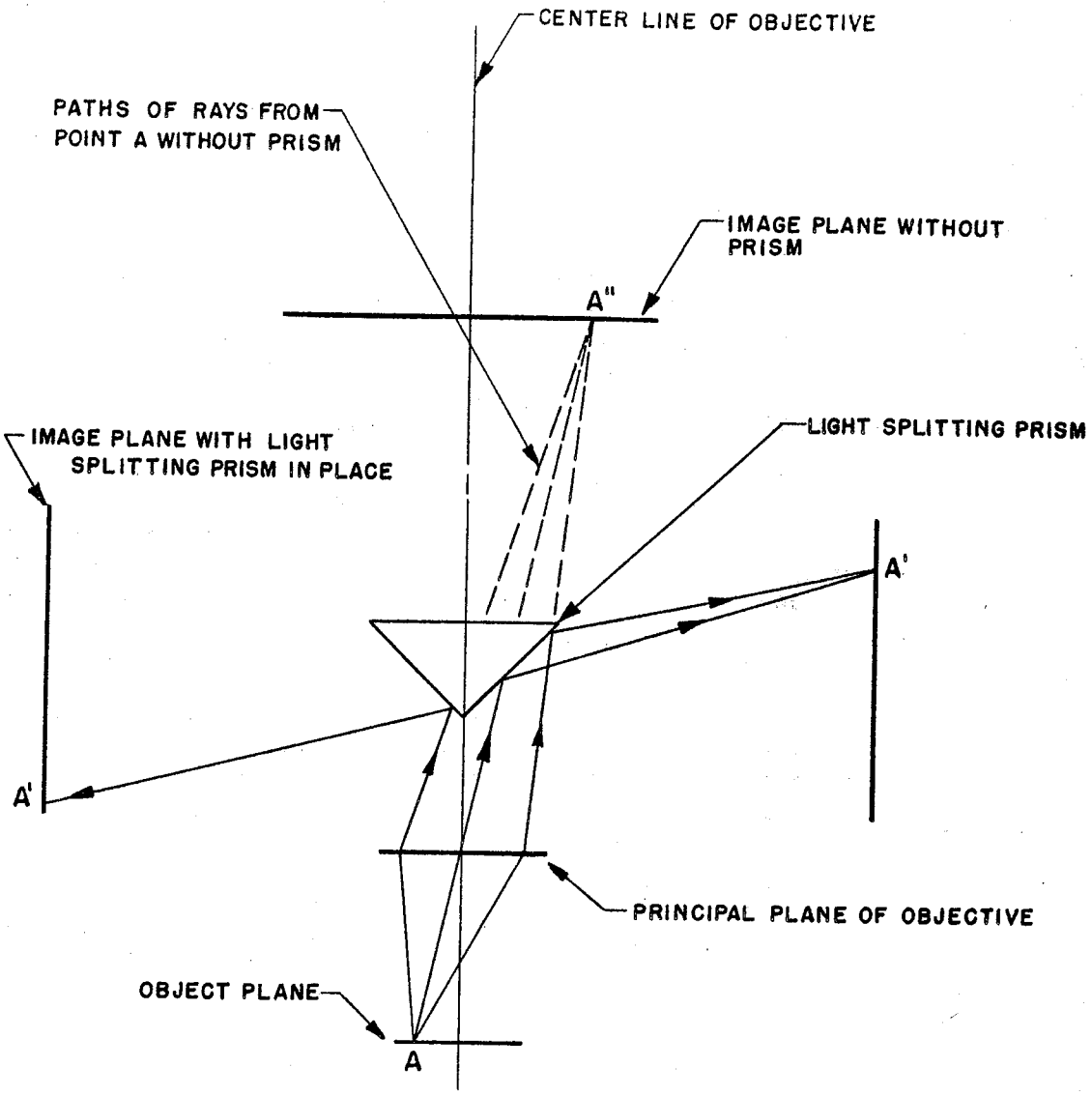

FIG. 2

SIMPLIFIED DIAGRAM OF PATH OF LIGHT THROUGH OPTICAL SYSTEM CONSISTING OF OBJECTIVE AND LIGHT SPLITTER

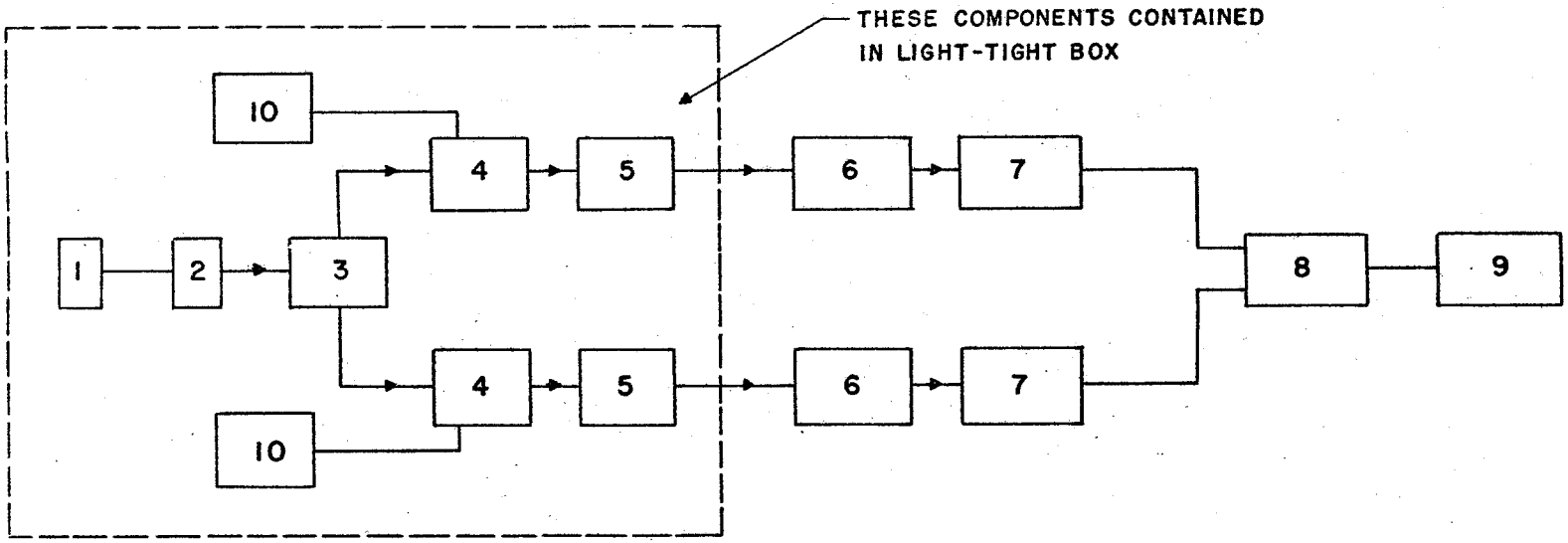

FIG. 3

BLOCK DIAGRAM OF APPARATUS

1 SOURCE
2 PHOSPHOR
3 OPTICAL SYSTEM
4 PHOTOTUBES
5 PULSE PREAMPLIFIER
6 PULSE AMPLIFIER AND SHAPER
7 PULSE AMPLITUDE SELECTOR
8 DISCRIMINATOR
9 RECORDER
10 H.V. SUPPLY FOR PHOTOTUBE

FIG. 4

MEASURED ACTIVITY VS DISTANCE FROM EDGE OF SOURCE

SOURCE Tl^{204} PLATED ON NICKEL-PLATED COPPER. ANTHRACENE CRYSTAL 250 MICRONS THICK. 97X OBJECTIVE. REAL FIELD 75 MICRONS