

SCINTILLATION COUNTING OF RADIATION  
USING ORGANIC COMPOUNDS

by

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## INTRODUCTION

A photoelectric scintillation counter is a device which detects radiation by making use of the fluorescence of certain materials and the high amplification factor of a photoelectric cell. A schematic diagram of such a counter as used by Westinghouse may be seen in Plate I. The fluorescent material may be in the form of a screen or a large transparent crystal. Work has been done using both forms of the fluorescent materials (1, 2). For gamma ray counting, a large transparent crystal is more efficient as these crystals are capable of absorbing most of the incoming radiation. The resultant fluorescent light, even though it originates inside the crystal, can be optically focused on the photo-surface of the multiplier cell.

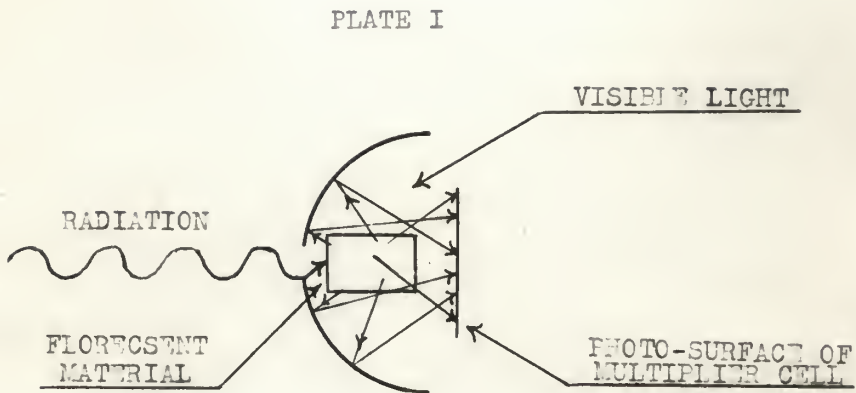
The method of photoelectric scintillation counting using naphthalene crystals, which must be cooled in order that their maximum efficiency may be obtained, was first used by Kallman (3) and later by Deutsch (4). Anthracene, suggested by both of these men as a fluorescent material, has the advantage over naphthalene in that its fluorescence is nearly independent of temperature (5); thus it may be used efficiently at room temperature.

Some of the advantages of a scintillation counter are:

1. A high gamma ray efficiency (1)
2. A short response time (1)
3. A high adaptability (1)

EXPLANATION OF PLATE I

Schematic diagram of a photoelectric scintillation counter.



4. A high sensitivity (1)
5. A relatively low cost.

The purpose of this research problem was to grow large, transparent, anthracene crystals for use with a photoelectric multiplier cell as a radiation detector.

## APPARATUS

### Purification Apparatus

The apparatus used in the purification of anthracene, with the exception of a suitable distillation flask and condenser, was obtained from the ordinary stock at hand. A satisfactory distillation flask and condenser were made from a short neck, five hundred milliliter, round bottom flask. Thirty millimeter pyrex tubing was fused to the neck of the flask and bent so the angle between it and the tube was approximately forty five degrees. This addition to the flask formed an air cooled condenser tube which was later cut to a length of twenty inches.

### Lowering Apparatus

In growing anthracene crystals, it is necessary, during crystal growth, to slowly lower the container through the furnaces. To accomplish this a motor with a built-in set of

reduction gears was connected to a worm gear which further reduced the rate of rotation by a ratio of seventy five to one. The output shaft of this reduction system was  $3/32$  inches in diameter. A single layer of soft copper wire was wound on this shaft, passed over a pulley and attached to the anthracene container. This apparatus is shown in Plate II.

It was necessary to attach a weight to the copper wire to keep it unwinding uniformly as the shaft rotated. Spreading of the turns of copper wire during operation, which would change the lowering rate of the sample, was avoided by holding the turns in place with an adjustable compression spring.

This apparatus lowered the anthracene tube at an approximate rate of  $1/7$  inch per hour.

#### Furnaces and Temperature Measuring Apparatus

Two separate furnaces, placed in a vertical position and separated by a sheet of tin foil approximately 0.3 mil thick, were used to control the growth of the anthracene crystals. The upper furnace was 18 inches in length and the lower furnace was 12 inches.

The heating coils of the two furnaces were made of nichrome wire wound on a four inch alundum core. A larger alundum core was placed around the windings for insulation and the two cores were fastened to the ends of a rectangular transite case by means of a transite collar. Asbestos wool was used for insulation around the outer core.

EXPLANATION OF PLATE II

Photograph of crystal growing apparatus

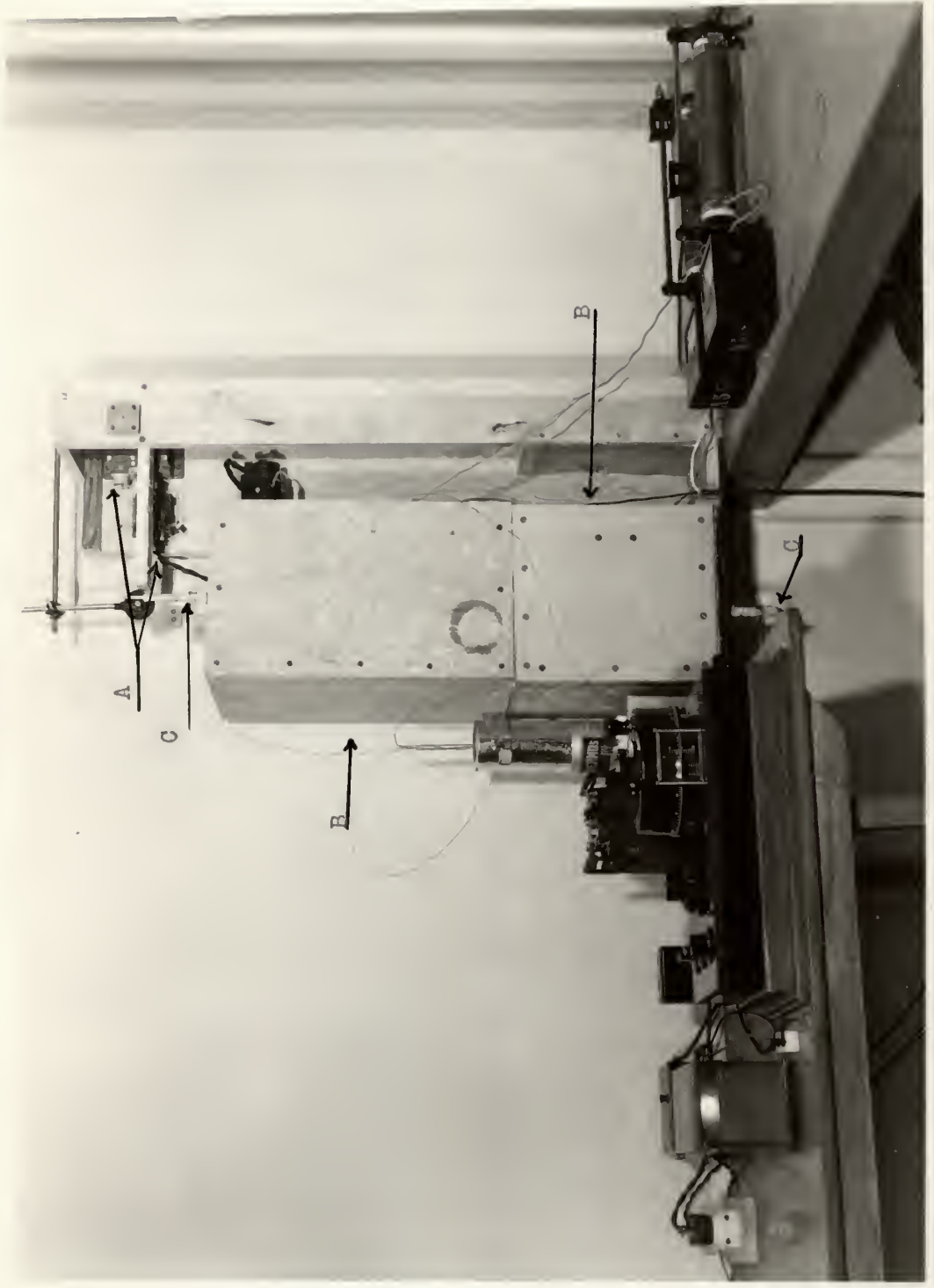
A - Lowering apparatus

B - Furnaces

C - Thermocouples



PLATE II



Chromel P-alumel thermocouples were inserted in each furnace as shown in Plate III. Each thermocouple was held one half inch from the walls of the furnace by threading it through a length of quarter inch brass tubing. The junction of the thermocouple was thermally insulated from the brass tubing with a two inch length of transite. Each thermocouple holder could be moved along its vertical axis and held at any desired position by brass friction clamps attached to the ends of the furnace. Frequent reading of the thermocouples, and adjustment of the current through the furnaces, enabled one to control the temperature of the furnaces quite accurately.

#### EXPERIMENTAL PROCEDURE

##### Purification of Anthracene

Several methods for the purification of anthracene were used. These methods were:

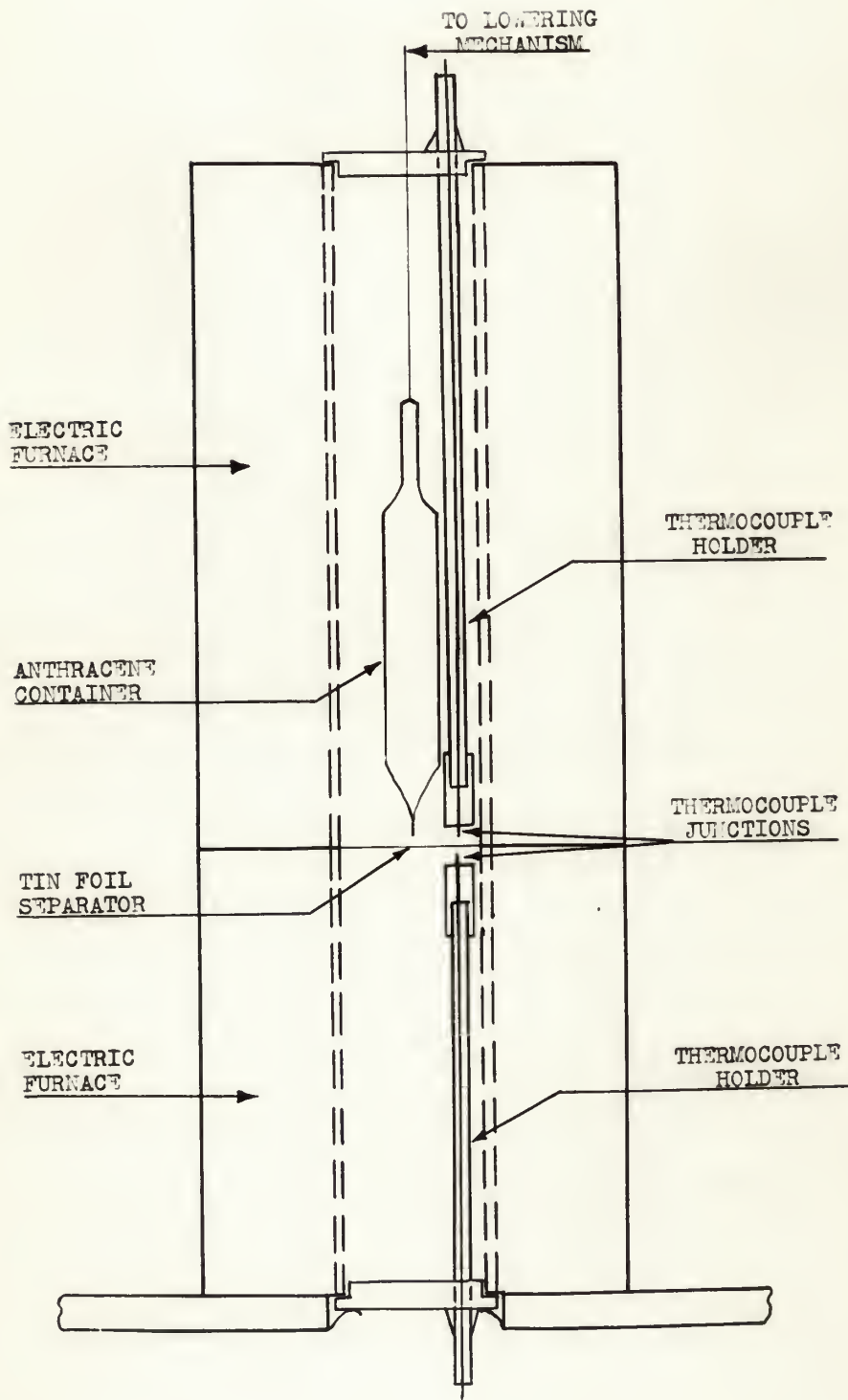
1. recrystallization with benzene (1)
2. extraction with acetone (7)
3. sublimation in vacuo (7)
4. codistillation with chemically pure ethylene glycol (7, 8).

Preliminary experiments involved repeated recrystallization of commercial anthracene with benzene. The benzene and anthracene were combined to near saturation at the boiling point of

EXPLANATION OF PLATE III

Schematic diagram of crystal growing apparatus.

## PLATE III



benzene, then filtered, while hot, in a Buchner funnel.

Later chemically pure anthracene was obtained from the A. D. Mackay Company and was further purified by several methods, as described in the following paragraphs.

In the extraction process anthracene and acetone were mixed in the ratio of four milliliters of acetone to one gram of anthracene. This mixture was shaken vigorously for several minutes, filtered through a Buchner funnel, removed from the funnel, and again shaken with a similar quantity of fresh acetone. After repeating the filtering process, the anthracene was allowed to dry while spread on filter paper.

A third purification process tested was that of sublimation under a forepump vacuum of approximately  $10^{-3}$  millimeters of mercury. The anthracene was heated to about  $200^{\circ}$  C. and was allowed to sublime into a two liter air cooled flask.

The anthracene for the last of the crystals grown was purified by extraction with acetone and codistillation with ethylene glycol using the flask and condenser previously described. It was then washed several times with hot distilled water, partially dried in a Buchner funnel, removed from the funnel and placed in a furnace at about  $70^{\circ}$  C. until thoroughly dry.

One other method was tested. Anthracene, which had been extracted with acetone, was recrystallized with benzene. In order to increase the percentage of recovery of anthracene, the Buchner funnel was cooled with an ice pack during this recrystallization process.

### Melting Point Determination

The purity of the anthracene was tested by a determination of the melting point of the compound. Pure anthracene melts at 217° C. (6) with impurities tending to lower the melting point. All the melting point determinations were made by placing powdered anthracene in a thin walled capillary tube which was fastened by a rubber band to the bulb of a mercury thermometer. The thermometer and capillary tube were heated in a mineral oil bath which was stirred during the experiment. Zero and exposed column corrections were made for the thermometer (9).

### Method of Growing Anthracene Crystals

Crystals may be grown from a vapor, a melt, a solution, or a solid (10). Of these four methods it was decided the most feasible method of growing anthracene crystals would be from the melt (1, 7, 8, 10). The crystals were grown in a pyrex tube having a diameter of twenty eight millimeters and an overall length of approximately twenty five centimeters. This tube is shown in Plate III.

The tube was filled with anthracene, evacuated with a forepump to approximately  $10^{-3}$  millimeters of mercury and then sealed.

The tube was placed in the upper furnace which was then adjusted to approximately 218° C. Simultaneously the lower

furnace was adjusted to approximately 207° C. The lowering apparatus was engaged, and these temperatures were maintained throughout the lowering process.

As the tube was slowly lowered, it punctured the tin foil separation, decreasing the temperature of the lower portion of the anthracene below its melting point, so that a crystal could form.

After the crystal was formed, it was necessary to cool it very slowly to reduce the strains, thus minimizing cracking.

During the cooling process, it was necessary to maintain the upper portion of the tube at a slightly higher temperature than the lower portion, so that the anthracene would not sublime from the top of the crystal.

The crystal was removed from the tube by opening the glass envelope. The good sections of the crystal were removed from the solid mass by cutting along the crystal boundaries with a razor blade. These pieces were shaped by grinding with numbers 500 and 2f crystalline alumina. Jewelers rouge was used for the preliminary polishing, after which the final polishing was accomplished with a cloth soaked in benzene.

#### EXPERIMENTAL RESULTS

The results of the various purification methods tested are shown in Table 1. The method of purification by sublimation under a vacuum was discarded because of the difficulties

encountered in removing the sublimed anthracene, in maintaining the seal of the ground glass joints at 200° C., and in preventing the anthracene from plugging the tube connecting the anthracene container to the receiving flask.

Table 1. Results of the various purification methods tested.

Method of purification :	Purifying agent :	No. times process was repeated on sample :	Melting point after purifying* :	Percent of recovery :
Recrystallization	Benzene	2	185	
Recrystallization	Benzene	5	216.9	Less than 10
Extraction	Acetone	2	215.2	81
Extraction and codistillation	Acetone	2	216.3	55
	Eythlene glycol	1		
Extraction and recrystallization	Acetone	2	215.8	62
	Benzene	1		
Sublimation				Unsatisfactory

\*The melting point determinations have an estimated accuracy of 1° C.

The crystals obtained from the first attempt at crystallization, in which purification of the anthracene was by extrac-



tion with acetone, were few in number, rather small and had a slight brown tinge which lowered their transparency.

About a dozen crystals were obtained from the last trial, in which the purification of the anthracene was by extraction with acetone and codistillation with ethylene glycol, the largest being 5 x 2 x .7 centimeters. These colorless, transparent crystals are shown in Plates IV and V.

The improvement shown in the results of the last trial may be due to the combination of six factors. First, purification of the anthracene by codistillation may have not only increased its purity, but possibly removed the coloring material. Second, the diameter of the tube was increased from 21 to 28 centimeters. Third, the quantity of anthracene used was increased from 32 grams to about 60 grams, thus increasing the possibility of more and larger crystals. Fourth, the lowering rate was decreased and made more uniform for the last trial. Fifth, the cooling period was increased from seventy to ninety six hours, thus reducing the cracking. Sixth, the anthracene tube for the last trial was evacuated for twelve hours, during which time outgassing was aided by placing it in a furnace at about 70° C. for four hours. This increased evacuation probably reduced oxidation during crystal growth.

EXPLANATION OF PLATE IV

Anthracene crystals as removed from furnace.

## PLATE IV



EXPLANATION OF PLATE V

Fig. 1 - anthracene crystals before polishing.

Fig. 2 - anthracene crystals after polishing.

## PLATE V



Fig. 1

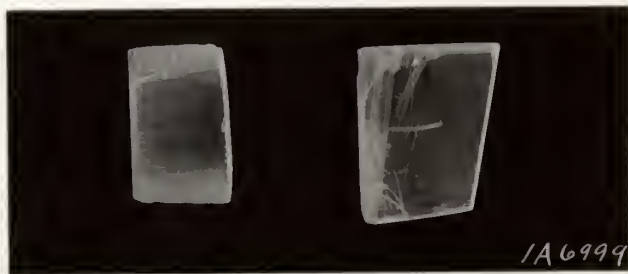


Fig. 2

## CONCLUSION

Of the several methods of purification, the most satisfactory was extraction with acetone combined with codistillation with ethylene glycol.

Transparent crystals of suitable size to be used for photoelectric scintillation counting were grown. The apparatus and techniques used in the purification of the anthracene and in growing the crystals were quite satisfactory; however, there are several suggested changes in procedure which could possibly result in even larger crystals.

The impurities in the anthracene are found in two places in the crystal tube; either in the bottom of the tube or at the boundaries between the crystals. According to Davey (10), impurities form at the crystal boundaries when the rate of cooling is too rapid. Therefore, it is suggested that the cooling rate be decreased. Further purification to remove these impurities would also be helpful. This might possibly be accomplished by repeated melting and recrystallization under a vacuum, since the larger portion of the impurities were always found at the bottom of the crystal tube.

Growing the crystals under high pressure of an inert gas should be tried in an effort to increase their size.

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