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MEASUREMENT

BY

J. SHARPE

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# NUCLEAR RADIATION MEASUREMENT

*by*

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## *Preface*

The subject of detection of nuclear radiation lends itself to a variety of treatments, depending on the space available. In the short compass of this monograph attention has been concentrated on the basic physical operation of the detectors and comparatively little attention has been paid to technical aspects of construction, although one chapter has been given over to a short survey of the associated electronics equipment necessary for efficient operation.

The treatment is based in part on articles which have appeared in *Nuclear Engineering* and I am grateful for the facilities given by the Publishers of that journal to reproduce tables and illustrations. The sections dealing with photomultiplier tubes and scintillation counters have been discussed at various times with my colleagues, Dr. E. E. Thomson of E.M.I. Electronics Ltd. and Mr. R. B. Owen of the Atomic Energy Research Establishment, Harwell.

Permission to contribute to this series of monographs was kindly afforded me by the Directors of Methuen and Co., Ltd., and I am grateful to the Directors of E.M.I. Electronics Ltd. for allowing me to undertake the project.

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J. SHARPE

June 1959



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## *Chapter One*

### INTRODUCTION

Elementary particles and high-energy quanta are not directly perceptible by man in sub-lethal doses, and instruments of varying types have been devised to record their presence and provide quantitative information relating to their energy, number and quality.

These detecting instruments are of two general sorts, passive elements, which store up information as long as they are exposed to the radiation and which have to be inspected in order to extract the data (e.g., a photographic emulsion), and active elements which produce an electrical signal indicative of the presence of radiation (e.g., a Geiger counter) but, whatever the mode of operation of the detector itself, the first requirement is that the nuclear radiation shall dissipate some of its energy within the sensitive element. This energy, which originally resided in the single primary particle, is then spread out over an appreciable volume of the detection medium and gives rise to events on a macroscopic scale which enables an observation to be made. In some cases, the useful events give rise to ionization, and the separation of the positive and negative ions by an electric field produces a signal which can be amplified and recorded. In other cases, the particle energy is used to produce many light quanta, which can be recorded by the intermediary of a photoelectric cell. In many passive detectors, the requirement is the initiation of a permanent chemical or physical change, such as takes place in the small crystals of silver halide in a photographic emulsion, or in some chemical dosimeters.

Nuclear radiation consists of three general types. Charged particles, such as electrons,  $\alpha$ -particles, protons, etc.; uncharged particles, such as neutrons and neutrinos, and electromagnetic radiation, such as  $\gamma$ -rays and X-rays. The sharing-out of energy in matter is done by interaction with the electronic structure, and so is only effective with charged particles, and neutrons and  $\gamma$ -rays must first interact in a special way to produce a charged particle before they can be detected. Before discussing detectors themselves in detail then, it is necessary to consider the ways by which interaction can take place and relate these to the various detection media available. This will be the subject of Chapter Two.

Once a particle has spent some of its energy within a detector the sequence of events is generally not dependent on the nature of the initial source of energy, so that detectors can be considered in their

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basic form before discussing the modifications required for use under particular conditions, and this is the task of Chapter Three, which deals with the fundamental theory of such devices as ion-chambers, Geiger counters, scintillation counters and cloud chambers.

Most of the remainder of the book then considers the specific problems raised by different types of radiation, and deals with some of the technical aspects of detectors.

Within the space available, it is not possible to consider in detail the electronic instruments which are used in conjunction with detectors, but a short Chapter Four relates the characteristics of detectors to those of the electronics.

## Chapter Two

# INTERACTION OF RADIATION WITH MATTER

## EQUIVALENCE OF MASS AND ENERGY

Before dealing with the manner in which interaction with matter takes place, it will be useful to summarize the data relating to the fundamental particles and the units of mass and energy.

The special theory of relativity provides a relationship between mass and energy,  $E = mc^2$  where  $c$  is the velocity of light, and  $m$  is related to the rest mass  $m_0$  by  $m = m_0/\sqrt{1-\beta^2}$  where  $\beta = v/c$  ( $v$  = particle velocity). Thus, the rest mass of an electron can be expressed in terms of energy, and a convenient unit for this purpose is  $10^6$  electron volts, ( $= 1 \text{ MeV} = 1.6 \times 10^{-6}$  ergs). In these units,  $m_0c^2$  for an electron or positron is 0.51 MeV; a proton is 936 MeV; an  $\alpha$ -particle, 3718.4 MeV; a neutron, 937.3 MeV; a  $\mu$ -meson 106 MeV; and  $\pi$ -mesons 135–139 MeV. The values of charge, in multiples of the electron charge,  $e = 1.6 \times 10^{-19}$  coulombs, are, positron  $+e$ ; proton  $+e$ ;  $\alpha$ -particle  $+2e$ ;  $\mu$ -meson  $+e$ ;  $\pi$ -mesons, 0 or  $\pm e$ ; neutron, 0.

Of the particles listed, those of least practical importance in the field of nuclear engineering are the mesons, and so, apart from considering them as a component of the background radiation, no special description of their properties will be given. Of the others, it will be seen that the neutron is different in having no charge, although having a mass as great as the proton.

## LOSS OF ENERGY IN MATTER

As a medium-energy charged particle, such as a 5 MeV proton, passes through matter it will lose energy mainly to the electrons of the atoms close to its track, by a series of inelastic interactions, and it can be seen intuitively that the rate of loss of energy by this means will become larger as the particle is slowed down, so giving more time for the transfer of energy to any given electron.

In elementary material of atomic number  $Z$ , atomic weight  $A$  and density  $\rho$ , the number of electrons per gramme is  $L.Z/A$ , where  $L$  is Avogadro's number,  $6 \times 10^{23}$ . The total cross-section of electrons per  $\text{cm}^2$  is then  $\pi r_0^2 L.Z/A$  per  $\text{gm} \cdot \text{cm}^{-2}$  or  $\pi r_0^2 L \rho Z/A$  per cm, where  $r_0$  is the classical electron radius,  $e^2/m_0c^2$  ( $\pi r_0^2 \cdot L = 0.154$ ), and the rate of energy loss is proportional to this factor. The electrons within the radius of action of the electrostatic field of the charged particle will be



“plucked” as it passes, so slowing it down, and the energy thus gained will be expended in various transitions in the electron shell of each atom, some of which will result in ionization. The specific energy loss, i.e. the energy loss per gm.cm<sup>-2</sup>, for a particle of velocity  $\beta c$  and mass  $M$  has been calculated by Bethe<sup>1</sup> as

$$-\frac{dE}{dx} = 4\pi\epsilon_0^2 L \frac{Z}{A} \cdot \frac{m_0 c^2 \cdot z^2}{\beta^2} \left[ \log \frac{2m_0 c^2 \beta^2}{ZI(1-\beta^2)} - \beta^2 \right]$$

Here  $ZI$  is an average excitation energy per atom,  $I$  being about  $11.5 \times 10^{-6}$  MeV, and  $ze$  is the charge on the particle. Putting  $\pi\epsilon_0^2 L = 0.154$  and  $m_0 c^2 = 0.51$  MeV, we have

$$-\frac{dE}{dx} = 0.314 \frac{Zz^2}{A\beta^2} \left[ \log \frac{1.02\beta^2}{ZI(1-\beta^2)} - \beta^2 \right]$$

For particles of  $\beta \ll 1$  and of mass  $M$ , this becomes

$$-\frac{dE}{dx} = 2\pi e^4 L \frac{Z}{A} \cdot \frac{z^2}{E} \frac{M}{m_0} \log \left( \frac{4m_0}{M} \cdot \frac{E}{ZI} \right)$$

Thus, an  $\alpha$ -particle having four times the energy of a proton, will lose energy four times as fast, and the two particles will have the same range except at low energies when capture and loss of electrons reduces the proton range by 3 mm in air compared with an  $\alpha$ -particle. The range of the particle,  $\int_0^E dE/(dE/dx)$  is approximately proportional to  $E^{1.8}/z^2 \cdot M^{0.8}$  (as compared with even the more approximate Geiger relationship  $R \propto E^{3/2}$ ).<sup>2</sup>

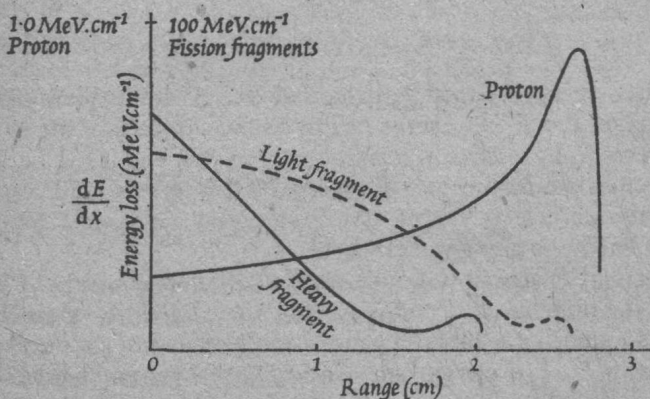


FIG. 1. Rate of loss of energy of proton (energy 1 MeV) and fission fragments, in argon. 760 mm Hg 15°C

Figure 1 shows  $dE/dx$  along the track of a 1 MeV proton in argon, together with curves for fission fragments which have masses of 98,000 and 148,000 MeV. It will be seen that these latter do not show the typical Bragg curve, with maximum energy loss at the lowest energy. This is because they are formed in fission with  $z \simeq 25$ , and capture electrons from the material around as they slow down, which reduces the rate of energy loss. Table 1 (overleaf) gives values of range for various particles in different materials.

At low energy, heavy particles may suffer elastic collision with a nucleus, and a process of this kind causes the increase in energy loss at the end of the track of fission fragments, as shown in Fig. 1. At high energy, the relativistic terms in the Bethe formula give rise to an increase in energy loss; the minimum rate of loss is found for particles of  $\beta = 0.98$  at which, in material of medium atomic number, singly charged particles will lose approximately  $2 \text{ MeV/g.cm}^{-2}$ .

As the velocity rises above  $\beta = 0.98$ , the increase in specific energy loss is found to be less than that predicted by Bethe, particularly in materials such as carbon, with a high electron density.<sup>3</sup>

The equation describing the rate of energy loss by electrons is slightly different from that given above, due to the identity of the two particles involved in the electron-electron interaction, but the chief difference from the case of heavy particles is due to the high degree of scattering experienced by electrons. As a result, the track of any individual electron may deviate very considerably from a straight line, and if the number of electrons in an initially collimated beam is plotted against range, an approximately exponential absorption will be found. The electrons from radioactive isotopes further complicate the picture by being emitted in a continuous spectrum up to a high energy limit  $E_{\max}$ , which is characteristic of the material. In fact, however, the range for these  $\beta$ -rays is not very different from that found for monoenergetic electrons of the same maximum energy and two empirical equations have been found to connect the range  $R$  in aluminium with  $E_{\max}$ , namely

$$0.1 < E_{\max} < 0.8 \text{ MeV} \quad R (\text{mg.cm}^{-2}\text{Al}) = 407E_{\max}^{1.38} \quad (E \text{ in MeV})$$

$$0.8 < E_{\max} < 2.5 \text{ MeV} \quad R (\text{mg.cm}^{-2}\text{Al}) = 541(E_{\max} - 0.245)$$

(Relations of the second type are called Feather equations.)

Owing to the exponential absorption of  $\beta$ -particles, a half-value absorber thickness may be found which is related to  $E_{\max}$ , by the formula  $a_{1/2} \sim 32E_{\max}^{1.38} (a \text{ mg.cm}^{-2} \text{ Al}, E_{\max} \text{ MeV})$  and in general the half-value thickness will be found to be approximately one-seventh of  $R_{\max}$ .

Table 1 summarizes some of the data relating to  $\beta$ -rays. The positive charge on a positron results in a different degree of scattering,

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TABLE 1. RANGE OF THE  $\alpha$ -PARTICLES, ELECTRONS AND FISSION FRAGMENTS IN VARIOUS MATERIALS

$\alpha$ -particles					Electrons	
Energy (MeV)	Air* (cm)	H <sub>2</sub> * (cm)	A* (cm)	Al (mg. cm <sup>-2</sup> )	Air* (cm)	Al (mg. cm <sup>-2</sup> )
0.01	—	—	—	—	0.22	0.27
0.05	—	—	—	—	3.6	5
0.1	0.1	0.3	—	—	11.2	13
0.5	0.3	0.88	—	0.5	145	170
1.0	0.5	1.9	0.5	0.8	370	405
2.0	1.0	3.9	1.1	1.6	830	926
4.0	2.5	11	2.8	—	—	—
5.3	3.8	17.6	4.2	5.8	—	—
7.0	5.9	27.8	6.4	—	—	—
10	10.6	51.5	10.9	—	—	5,200

Fission fragments: light,  $R$ , (mg. cm<sup>-2</sup> Al) = 4.25  
heavy,  $R$ , (mg. cm<sup>-2</sup> Al) = 3.6

\* 760 mm Hg, 15°C.

but the range in aluminium is about the same as for electrons. A greater range is found in materials of high  $Z$ , however.

At the end of its range a positron finds itself a lone positive particle in a sea of negative electrons, and electrostatic forces result in its combination with an electron to give two quanta of  $\gamma$ -rays of energy 0.51 MeV (annihilation radiation), the presence of which is characteristic of the emission of positrons.

## DETECTION MEDIA

So far we have considered the loss of energy from charged particles without inquiring into the form in which this is distributed in the detection medium, other than by the excitation of electrons. In some cases, sufficient energy will be communicated to one of these so that it leaves its parent atom with sufficient velocity to produce excitation and ionization on its own account, and about half of the ionization in a gas produced by a fast particle, such as a high-energy electron or a meson, is produced by the intermediary of these  $\delta$ -rays.

It is obvious that only a proportion of the interactions with electrons will result in the ionization of an atom or molecule, with the production of a free electron and a positive ion, and that in the other cases, excitation

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short of ionization will result in radiation of light or the production of thermal energy as the excited structure returns to its ground state. In some materials, such as gases, the production of ionization is generally the desired end, while in others, a high efficiency of light production is required, while in still others chemical change or temperature rise is wanted.

### IONIZATION MEDIA

#### Gases

By measuring the number of ions produced in a gas by absorption of a known amount of energy  $E$  from a particle, the value of  $w$ , the energy per ion pair, can be found, and some values are given in Table 2,

TABLE 2. IONIZATION POTENTIAL,  $E_i$ , AND ENERGY PER ION PAIR,  $w$ , FOR VARIOUS GASES

Gas:	Argon + 0.2% N <sub>2</sub>	Argon + Ben- zene	He	He + 0.13% argon	Ne	Ne + 0.12% argon	Kr	Xe	Air	H <sub>2</sub>	BF <sub>3</sub>	CH <sub>4</sub>
$E_i$ (eV):	15.7		24.5		21.5		13.9	12.1	16.7 & 12.8*	15.9	10.3	15.2
$w$ (eV) for 5.3 MeV alpha:	26.3	22.9	41.3	29.7	36.3	26.1	26.0	23.6	35.6	36	35.2	30.0
$w$ (eV) for H <sup>+</sup> betas:	28.5								31.0			28.5

Argon + 0.2% N<sub>2</sub>,  $w$  for: Fission fragments, (light) 27.6 eV, (heavy) 29.4 eV. Recoil nucleus from Po decay, 70 eV.

\* 12.8 for O<sub>2</sub>, 16.7 for N<sub>2</sub>.

together with values of the ionization potential,  $E_i$ , of the particular gas. It will be noted that mixtures of gases may give more ionization than a pure gas, and this is due to the fruitful use of some of the excitation energy in a gas of high ionization potential passed on by collision to a gas of lower  $E_i$ . The decreasing efficiency of ionization of very slow particles will also be noted, and is to be expected from the occurrence of elastic collisions which produce recoil atoms too low in energy to produce ionization.

The initial ionization will consist of electrons and positive ions, which may be separated under the influence of an electric field to prevent recombination. In some gases, the electrons remain free and will move with high mobility towards the positive electrode, while the much

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more massive positive ions move slowly towards the cathode. In other gases, however, electrons are rapidly attached to neutral molecules, to form negative ions, and then have a low mobility similar to positive

TABLE 3. MOBILITIES OF POSITIVE IONS AND ELECTRONS IN GASES

Gas	Ion	$\mu_+$ ( $\text{cm}^2 \cdot \text{V}^{-1} \text{sec}^{-1}$ )	$\mu_-^*$ ( $\text{cm}^2/\text{V}^{1/2} \text{sec}^{-1}$ )
He	$\text{He}^+$	10.8	$3.5 \times 10^4$
	$\text{He}_2^+$	19.0	
A	$\text{A}^+$	1.63	$7 \times 10^4$
	$\text{A}_2^+$	1.9	
A + 1% $\text{N}_2$			$8 \times 10^4$
A + 10% $\text{C}_2\text{H}_5\text{OH}$	$\text{C}_2\text{H}_5\text{OH}^+$	0.7	$6 \times 10^4$
$\text{N}_2$	$\text{N}_2^+$	2.7	$3 \times 10^4$
$\text{H}_2$	$\text{H}_2^+$	14.7	$5 \times 10^4$

\*  $W$  = velocity of electron =  $\mu_-(X/p)^{1/2}$  where  $X$  is electric field ( $\text{V} \cdot \text{cm}^{-1}$ ) and  $p$  is pressure.

ions. Table 3 gives data on free-electron gases, values of  $\mu_+$ , the positive ion mobility being defined as the velocity in  $\text{cm} \cdot \text{sec}^{-1}$  per volt  $\cdot \text{cm}^{-1}$  at one atmosphere pressure. The very approximate figures given for  $\mu_-$ , the electron mobility, are defined from the equation  $W$  (electron velocity) =  $\mu_-(X/p)^{1/2}$  where  $X$  is the electric field strength in volts per cm and  $p$ , the pressure, is measured in atmospheres. Figure 2 shows how the electron velocity varies in practice with  $X/p$  for various gases, and again, the influence of small admixtures to a noble gas, such as argon, will be noted. In this case, the effect is due to the reduction in the agitation energy of electrons by inelastic collision with the more complex molecules, and the consequent increase in mean free path. This is known as the Ramsauer effect.<sup>5</sup>

Some gases which form negative ions are  $\text{H}_2\text{O}$ ,  $\text{O}_2$ ,  $\text{SiF}_4$ , and the halogens. In the absence of an electric field, the positive ions and electrons will diffuse around their point of formation, and will either recombine directly, or after the electrons have formed negative ions. In weak fields, recombination will still occur, and is proportional to the ion density in the region. A recombination coefficient may be defined by  $-dn/dt = \alpha n_+ n_-$  where  $n_+$  and  $n_-$  are densities of positive and negative ions (or electrons), respectively. Since the density of ions at a given moment after formation is inversely proportional to their drift



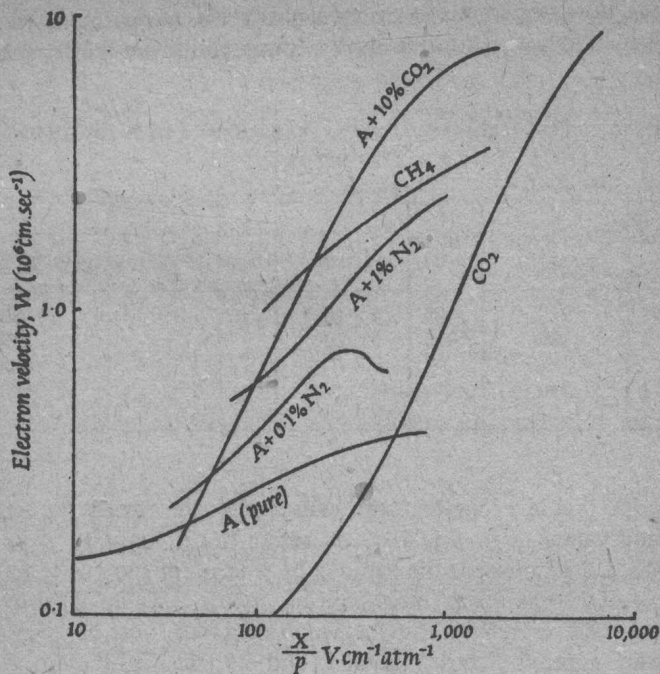


FIG. 2. Drift velocity of electrons,  $W$ , in various gases as a function of  $X/p$

velocity, recombination will proceed faster in low fields, and in gases which form negative ions, than in free electron gases at high values of  $X/p$ .

### Solids

Even in solids, ionization will occur on the passage of a charged particle and in most materials, such as metals, the free electrons produced will rapidly lose energy and cease to have an independent existence. In insulators and semiconductors with a perfect, or near perfect, crystal structure, free electrons will move through the material in the conduction band until trapped or recombined with an electron vacancy (or positive hole). Such free electrons will drift towards the positive electrode if a field is applied across the crystal and so may be used to produce a signal in an external circuit, just as is done with the ions in a gas chamber. The magnitude of the signal produced by a particular electron will depend on the distance travelled parallel to the field between the point of formation and the position in which the electron is

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trapped, and this is equal to  $S = \tau W$  where  $\tau$  is the mean free lifetime of the electron and  $W$  is the drift velocity,  $= \mu_- X$  ( $\mu_-$  is the electron mobility).  $S/X = \tau \mu_-$  and is characteristic of a given material; this is

TABLE 4. DATA ON MATERIALS USEFUL FOR SOLID IONIZATION DETECTORS

Material	Temperature (°K)	$w$ (eV)	$S/X$ (cm <sup>2</sup> . V <sup>-1</sup> )	$\mu_+$ (cm <sup>2</sup> . V <sup>-1</sup> sec <sup>-1</sup> )	$\mu_-$ (cm <sup>2</sup> . V <sup>-1</sup> sec <sup>-1</sup> )
Diamond	290	7-25	$5 \times 10^{-4}$	> 1,200	> 400
CdS	290	5-15	$5 \times 10^{-5}$		30
AgCl	70	7	$3 \times 10^{-4}$	0	139
Ge	290	3		1,700	3,500

tabulated in Table 4 together with values of  $w$ , the energy per ionizing event, and values of  $\mu_-$  and  $\mu_+$ .  $\mu_+$  refers to the mobility of positive holes which drift towards the cathode by a series of successive replacements of electrons from neighbouring atoms. Table 4 also gives information on the temperature at which materials must be operated to obtain the required electron mobility and electrical insulation.

## Secondary Emitters

In some insulators, the  $\delta$ -rays produced by the passage of the charged particle may sometimes emerge from the surface of the material as secondary electrons, materials having a high value of  $\tau \mu$  tending to be good secondary emitters. Some values of the secondary emission

TABLE 5. DATA ON SECONDARY EMITTERS

( $\delta_{\max}$  is maximum value of secondary emission coefficient, at voltage  $V_{\max}$ .)

Material	$\delta_{\max}$	$V_{\max}$	Material	$\delta_{\max}$	$V_{\max}$
Al	0.97	300	BeO	5 to 10	400 to 800
Be	0.90	400	MgO	4 to 8	400 to 600
Cu	1.35	500	AgMgOCs	10	400
Cs	0.90	400	SbCs	12	500
Al <sub>2</sub> O <sub>3</sub>	2.6				

$\delta$  vs.  $V$  for AgMgOCs varies approximately as  $V/40$  initially

$\delta$  vs.  $V$  for SbCs varies approximately as  $0.2V^{0.7}$  initially.

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coefficient  $\delta$  (defined as number of secondaries produced per incident particle) are given in Table 5.  $\delta$  is a function of incident electron energy, rising from zero to a maximum value of between 1 and 12 at a voltage around 500–600 V, and then falling off again. These values are for secondaries leaving the surface struck by the particle.

### PHOSPHORS

Thus far, we have considered the use of the energy of a charged particle to produce ionization, but it has been obvious that since the energy per ion is two or three times the ionization energy of an atom (Table 2), the amount of energy used in producing excitation short of ionization is very appreciable and will appear as light under suitable conditions.

Materials giving a relatively high efficiency of conversion of particle energy to light are called phosphors and are of two general types, organic and inorganic.

#### *Organic Phosphors*

Transparent organic materials, such as anthracene, which have conjugated double bonds in their benzene ring structure, will dissipate a high proportion of excitation energy as light, the wavelength of which is dependent on the complexity of the material, as shown in Table 6.

TABLE 6

<i>Material</i>	<i>Emission spectrum</i>
Benzene	2,600–3,000 Å
Naphthalene	3,000–3,650
Anthracene	3,800–4,550
Naphthacene	4,500–6,500
Pentacene	Bands in red

The de-excitation times for the electronic structure of such materials are very small, so that fluorescence decay times of the order of  $10^{-8}$  sec are observed (Table 7 (a)).

Since, in general, the light from a phosphor is to be absorbed in a photoelectric surface to produce photoelectrons, and the most efficient surfaces have their highest sensitivity in the blue region of the spectrum, around 4,200 Å, phosphors for use in radiation detectors will in general

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be chosen to have a blue light output. Tables 7 (a), 7 (b) and 7 (c) give details of phosphors, and photocathodes, together with values of  $w$ , the energy abstracted from the particle to produce a photoelectron from a specified photosurface optically coupled to the phosphor.

Many organic phosphors may be grown into quite large single crystals, which are very useful in scintillation counters, and observation of these shows that energy introduced into the material at one point, by, say,  $\alpha$ -particles of short range, may be transmitted to remote points of the crystal by a species of resonant transfer, until radiation occurs. If a molecule emitting at a longer wavelength than the matrix is introduced, this will radiate any energy, received from the matrix, at its characteristic wavelength. The efficiency of this process is illustrated by the fact that the addition of about 1 per cent of anthracene to

TABLE 7 (a)

Phosphor	Photocathode	Photo-sensitivity ( $\mu\text{A} \cdot \text{lm}^{-1}$ )	Peak quantum efficiency (per cent)	$w$ for $\frac{1}{2}$ MeV electrons (eV/photo-electron)	Decay constant $\tau_1$ ( $\mu\text{sec}$ )
NaI-Tl (0.1% Tl)	SbCsO	60	15	250	250
	SbCs	25	10	370	
	Sb-Na-K-CS	150	20	180	
(0.01% Tl) ZnS-Ag	SbCsO	60	15	500	3,000 ( $\tau^{-n}$ law)
	SbCsO	60	15	250	
CsI-Tl	SbCsO	60	15	1,000– 2,000	1,100
LiI-Eu	SbCsO	60	15	1,000– 2,000	
Anthracene	SbCsO	60	15	500	32
Stilbene	SbCsO	60	15	1,200	6
Plastic (Polyvinyl toluene and terphenyl and 2.5 diphenyloxazole)	SbCsO	60	15	1,500	5
Liquid (Toluene & 5 g/litre p-terphenyl and 16 mg/litre POPOP)	SbCsO	60	15	1,500	2
Xe	SbCsO in quartz	60		1,000 gas 350 solid ( $\alpha$ -particles)	1 $\mu\text{sec}$