

obtained by integrating over all directions of the second counter. At  $\Phi=30^\circ$ , the free  $p$ - $p$  cross section (laboratory system) is  $13.2 \times 10^{-27}$  cm<sup>2</sup>/sterad. Three times this cross section is  $39.6 \times 10^{-27}$  cm<sup>2</sup>/sterad. This compares remarkably well with the cross section  $(39 \pm 4) \times 10^{-27}$  cm<sup>2</sup>/sterad obtained from lithium. It should be mentioned, however, that the vertical spread, observed when counter  $B$  is moved out of the plane of  $\mathbf{P}$  and  $\mathbf{P}'$ , is about 40 percent larger than that to be expected from the present interpretation of the horizontal spread

(Figs. 1 and 2). This aspect will be investigated more closely in further work.

These experiments are preliminary in nature but they show qualitative features which seem of interest to us. When improved and extended, they ought to be able to give direct information on the motion of the protons inside of the nucleus and on the transparency of nuclear matter for protons.

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## Decay Times, Fluorescent Efficiencies, and Energy Storage Properties for Various Substances with Gamma-Ray or Alpha-Particle Excitation\*†

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The results of measurements of the decay time of light flashes induced by gamma-ray quanta, or alpha-particles by means of pulse distribution curves for various crystals, powders, and solutions are presented. The decay of organic substances can be described essentially by a single exponential; in addition, a small more slowly decaying portion is present. The decay of inorganic crystals can be represented by a sum of exponentials with time constants in different ranges. The results support a previous explanation of the lower efficiency of alpha-particles in producing fluorescence when compared to gamma-rays.

Results on various activated inorganic crystals of integrated fluorescent light output under gamma-rays and peak height measurements under gamma-ray and alpha-particle bombardment are also presented. The relative integrated and peak height properties are not well correlated, neither are the responses to gamma-rays and alpha-particles. Such variations are associated with the different emission times of the crystals. It is found that CsBr and CsI activated with Tl are excellent crystals with respect to luminescent properties especially when volume is a factor. The capability of these crystals to store energy to be released later as light by light of longer wavelengths is also indicated. Very good storage and stimulation properties are shown only by NaCl crystals activated with AgCl.

### I.

THE duration of scintillation pulses when a fluorescent material is excited by a single gamma-ray, photon, or alpha-particle has been extensively investigated. The results described in this paper are obtained by a method similar to that described previously.<sup>1</sup> The peak voltages of the pulses produced by a photomultiplier tube receiving the light flashes from the fluorescent material were determined as a function of the resistance at the output of the tube. (The experimental arrangement will be described in a future publication.) For this purpose, pulse distribution curves were measured under equivalent geometrical conditions for different output resistors, and for each the characteristic cut-off voltage was determined. These cut-off voltages provide information on how the light given off in the flash is emitted as a function of time. If the light flash decays exponentially, the time constant of the decay can be de-

termined readily from the ratio of peak voltages for two different resistors.<sup>1</sup> Measurements were made on various single crystals, solutions, and powders using gamma-ray and alpha-particle radiation. The results of the relative pulse-height measurements are reported in Table I.

The analysis of these measurements show that for the organic substances the light flash decays almost exponentially. The time constants calculated under this assumption are given in Table II for various organic scintillators. They are average values obtained from the measurements with different resistors from 100K ohms downwards to 500 ohms and are listed only in those cases where the results could be closely approximated by a single exponential. They agree fairly well with time constants measured by different methods.<sup>2</sup> A closer

<sup>2</sup> Liebson, Bishop, and Elliot, *Phys. Rev.* **80**, 907 (1950); A. Lundby, *Phys. Rev.* **80**, 477 (1950); R. F. Post and N. S. Shiren, *Phys. Rev.* **78**, 80 (1950); G. G. Kelley and M. Goodrich, *Phys. Rev.* **77**, 138 (1950); Hofstadter, Liebson, and Elliot, *Phys. Rev.* **78**, 81 (1950); Elliot, Liebson, Meyers, and Ravilious, *Rev. Sci. Instr.* **21**, 631 (1950); G. T. Kelley, Oak Ridge National Laboratory Report 366 (1949); G. B. Collins, *Phys. Rev.* **74**, 1543 (1948); R. C. Sangster, Massachusetts Institute of Technology Technical Report No. 55 (1952).

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<sup>1</sup> Broser, Kallmann, and Reuber, *Z. Naturforsch.* **5a**, 79 (1950).

TABLE I. Ratios of pulse voltages for various resistors with respect to pulse voltage at 1 megohm.

Substance	500K	100K	50K	10K	5K	1K	0.5K
Alpha-particle irradiation							
Single crystals							
Anthracene	0.94	0.85	0.82	0.70	0.60	0.36	0.25
Naphthalene	0.94	0.77	0.69	0.47	0.39	...	...
Calcium tungstate	0.84	0.47	0.36	0.21	...	...	...
Potassium iodide	0.90	0.67	0.59	...	...	...	...
Stilbene	0.94	0.79	0.74	0.65	0.60	0.47	0.37
Powders							
ZnS (Long persistent, Ag)	0.88	0.58	0.48	0.31	0.24	0.12	0.08
ZnS (Short persistent, ZnCd)	0.90	0.57	0.45	0.24	0.17	0.08	0.05
ZnO	0.87	0.61	0.50	0.28	0.21	0.10	...
Gamma-ray irradiation							
Single crystals							
Anthracene	0.98	0.91	0.86	0.71	0.59	0.33	0.23
Naphthalene	0.94	0.77	0.69	0.46	0.34	...	...
Cadmium tungstate	0.83	0.50	0.39	0.26	...	...	...
Calcium tungstate	0.82	0.44	0.31	0.17	...	...	...
Potassium iodide	0.89	0.62	0.52	0.29	0.23	...	...
Stilbene	0.96	0.87	0.83	0.75	0.69	0.54	0.41
Solutions							
$P+p.T$ (3)*	0.95	0.85	0.81	0.73	0.69	0.55	0.44
$X+p.T$ (5)*	0.93	0.77	0.74	0.66	0.61	0.51	0.40
$X+m.T$ (200)*	0.94	0.76	0.71	0.61	0.56	0.42	0.32
$P+p.T$ (3) + $D$ (0.015)*	0.97	0.85	0.82	0.73	0.69	0.52	0.40
$P+D$ (0.7)*	0.94	0.80	0.75	0.66	0.60	0.44	0.32
$P+D$ (4)*	0.95	0.76	0.71	0.63	0.57	0.43	0.32
$P$ = Phenylcyclohexane	$m.T$ = $m$ -Terphenyl						
$p.T$ = $p$ -Terphenyl	$X$ = Xylene						
$D$ = Diphenylhexatriene	* Numbers indicate grams per liter						

study taking the peak voltage values with higher resistors into account indicates, however, that even in the organic substances a better fit to the experimental decay curve would be obtained by a pulse consisting mainly of a single exponential decay (about 95 percent of the emitted light) followed by a slowly decaying tail. The measurements show further that for the organic substances the time constants are only slightly shorter for alpha-particle excitation than for gamma-ray excitation.

This result together with measurements on the integrated intensity of solutions excited by both types of radiation offer another possibility of checking our previous explanation for the much smaller light producing efficiency with alpha-particle excitation than with gamma-ray excitation for equivalent amounts of absorbed energy. The measurements<sup>3</sup> show that the parameters in the equation which describes the dependence of the light output as a function of the solute concentration, are only slightly different for gamma-ray and alpha-particle excitation, whereas the light output is one-tenth or less in the case of alpha-particles for the same amount of absorbed energy. These parameters describe essentially the period of time in which the energy is transferred from the solvent to the solute

<sup>3</sup> M. Furst and H. Kallmann, Phys. Rev. **85**, 816 (1952).

molecule and is quenched in the solute molecule. Hence, all these measurements show that the time constants involved are only slightly changed under alpha-particle excitation whereas the efficiency is strongly changed.

This latter phenomenon was previously explained<sup>3</sup> by the assumption that the excited molecules themselves quench the excitation energy by "collisions" with each other and thus reduce the light emission. It is essential in this explanation that the quenching effect for emission by alpha-particle excitation does not stay constant but decreases during the quenching and emission processes since the concentration of excited molecules decreases by means of the quenching itself. Before the main part of the light is emitted, most of the quenching has already occurred. It follows that the time constants of the light flash produced by alpha-particles differ only slightly from those excited by gamma-radiation. The mechanism proposed by Birks<sup>4</sup> for crystals cannot account for time constants of the same order of magnitude for alpha-particle and gamma-ray excitation since he assumes a quenching agent which does not decay in the same degree as the excitation energy of the molecules. It is assumed, therefore, that the explanation given previously gives an adequate description of the light emission process produced by alpha-particles.

The measurements of pulse heights for inorganic powders presented in Table I show quite definitely that the light emission of these powders does not decay exponentially. Much of the light is emitted over a period of about one microsecond. However, if one attempts to represent the experimental decay curves for the inorganic crystals by a sum of exponential functions with different time constants, at least several time constants in the range below one microsecond and also some with a

TABLE II. Average time constants of various substances.<sup>a</sup>

Substance	Time constants (seconds)
Gamma-ray irradiation	
Anthracene	$(22.8 \pm 4.8^b) \times 10^{-9}$
Naphthalene	$(87.5 \pm 0.5) \times 10^{-9}$
Stilbene	$(6.9 \pm 1.7) \times 10^{-9}$
Phenylcyclohexane + $p$ -terphenyl (3 g/l)	$(5.4 \pm 2.3) \times 10^{-9}$
Xylene + $p$ -terphenyl (5 g/l)	$(6.5 \pm 2.7) \times 10^{-9}$
Xylene + $m$ -terphenyl (200 g/l)	$(8.8 \pm 3.0) \times 10^{-9}$
Phenylcyclohexane + diphenylhexatriene (0.7 g/l)	$(9.3 \pm 1.8) \times 10^{-9}$
Phenylcyclohexane + diphenylhexatriene (4 g/l)	$(8.2 \pm 2.0) \times 10^{-9}$
Phenylcyclohexane + $p$ -terphenyl (3 g/l) + diphenylhexatriene (0.015 g/l)	$(6.8 \pm 2.0) \times 10^{-9}$
Alpha-particle irradiation	
Anthracene	$(16.5 \pm 1.5) \times 10^{-9}$
Naphthalene	$(53 \pm 23) \times 10^{-9}$
Stilbene	$(7.2 \pm 2.9) \times 10^{-9}$

<sup>a</sup> These values are lower than those reported in reference 1 since the previous determinations employed longer circuit time constants, which result in larger time constants as a consequence of the decay not being a single exponential.

<sup>b</sup> Average deviations.

<sup>4</sup> J. B. Birks, Proc. Phys. Soc. (London) **A64**, 874 (1951); Phys. Rev. **84**, 364 (1951).

TABLE III. Fluorescent and stimulation properties of crystals.<sup>a</sup>

Crystal	Effective mass g	Emitted wavelength	Fluorescent intensity compared to anthracene of same mass	Stimulated intensity by 1 min u.v. <sup>b</sup>
NaCl+AgCl 1%	6.5	Ultraviolet emission	1.0	56
NaCl+AgCl 1%	6.5	Ultraviolet emission	1.8	27
NaCl+AgCl 5%	5.7	Ultraviolet emission	0.8	20
CsI+Tl #A	9.4	Blue-green emission	1.1	...
CsI+Tl #1	11	Blue-green emission	1.3	...
CsBr+Tl	11	.....	3.0	...
KCl+AgCl	>10	.....	<0.1	0
KBr+TlBr 1/2%	5.7	Near ultraviolet emission	1.7	~4
NaCl+CuCl 1%	5.7	.....	0.1	~1
LiF+AgCl	5.7	.....	0.02	~0
CaF <sub>2</sub>	2	Far ultraviolet emission	0.7	<1

<sup>a</sup> Relative fluorescent intensities for solutions have been published previously in reference 3.

<sup>b</sup> After 30 seconds.

<sup>c</sup> Crystal from cone portion of large crystal—Emission has a greater percentage in the short u.v. than sections from the top of the large crystal.

much longer period must be assumed. If the decay were a simple exponential, the pulse heights should decrease approximately proportionally to the output resistance. The slow decrease in pulse height at the lower values of resistance, however, indicates that very short time constants are also present in the light emission decay curve of some of these substances. It may be noted that the zinc sulfides under alpha-particle bombardment show a pulse height for an output resistance of 500 ohms (corresponding to a time constant of  $10^{-8}$  second) which amounts to 12 percent of that of the 1 megohm resistance. This pulse height is approximately the same as that with an anthracene crystal with a 1 megohm output resistance.

## II.

A number of different crystals, mostly obtained from the Harshaw Chemical Company, have been measured with respect to fluorescent efficiencies and energy storage properties<sup>5,6</sup> under gamma-radiation. In addition, the light flash intensities (peak heights) have been measured under both gamma-ray and alpha-particle excitations. The integrated intensity measurements employing a 1P28 photomultiplier were made essentially with equipment described previously in connection with measurements of solutions.<sup>7</sup> The peak height measurements were made using a 1P21 photomultiplier with a scaler and discriminator. Some measurements have been previously reported on the fluorescence of similar crystals, e.g.,<sup>8-10</sup> but the comparison of various results and of storage properties have not been shown.

The integrated intensity properties for gamma-rays are presented in Table III. The fluorescent intensities are compared to an anthracene crystal of the same mass.

<sup>5</sup> M. Furst and H. Kallmann, *Phys. Rev.* **82**, 964 (1951).

<sup>6</sup> H. Kallmann and M. Furst, *Phys. Rev.* **83**, 674 (1951).

<sup>7</sup> H. Kallmann and M. Furst, *Phys. Rev.* **79**, 857 (1950).

<sup>8</sup> O. Glasser and I. E. Beaseley, *Phys. Rev.* **47**, 570, 789 (1935).

<sup>9</sup> C. E. Mandeville and H. O. Albrecht, *Phys. Rev.* **80**, 299 (1950).

<sup>10</sup> W. Van Sciver and R. Hofstadter, *Phys. Rev.* **84**, 1062 (1951).

This table shows that some of the crystals have a noticeably greater fluorescent efficiency than an anthracene crystal. In particular, these are cesium bromide and cesium iodide activated with thallium, and KBr activated with thallium bromide. These results essentially refer to the same amount of absorbed energy because this is almost proportional to the mass of absorbing material for hard gamma-rays. Considered from the point of view of equal volumes, cesium bromide and iodide are distinctly superior to anthracene because of their greater density. Since in radiation work using fluorescent substances the volume of crystal may be a limiting factor, these crystals are among the most effective for the measurement of weak high energy radiation. These crystals may also be valuable for visual observation since their emission spectra are partially in the green.

The intensities which can be stimulated by light (3600A) after excitation by gamma-radiation as a consequence of stored energy are considerable only for NaCl crystals activated with AgCl.<sup>5,6</sup> For other substances the stimulated intensities are very much smaller but easily observable. Since only a rather small dosage of gamma-rays were used in these experiments (of the order of 1 roentgen), the comparatively small amount of energy storage in the other crystals is only partly evident but some is present. Also in those cases where some storage is found with other crystals, the decay time of this stored energy is very much shorter than that of NaCl. NaCl+AgCl 1 percent crystals taken from different sections of a large crystal appear to show somewhat different properties. Thus crystals from the cone portion of the large crystal appear to have a greater percentage of emission in the shorter u.v. wavelengths and a greater fluorescence, but less storage ability.

With organic crystals, there is usually a rather high correlation between the relative peak heights and the total (integrated) light output for various substances. For the crystals herein reported, such a close correlation occurs in only a limited number of crystals as can be seen by comparing Table IV with Table III. The relative efficiencies for gamma-ray and alpha-particle are also not proportional for these crystals as can be seen from Table IV. The maximum pulse heights are corrected to a count rate of zero, and the partial destruction especially of organic crystals under prolonged alpha-excitation was taken into account in the measurements.

The pulse-height distribution for alpha-particle excitation using a 1 megohm resistor at the output of the photomultiplier is shown in Fig. 1 for cesium iodide and potassium iodide with a photomultiplier potentials of 1400 and 1600 volts, respectively. The cesium iodide has considerably greater peaks than the potassium iodide despite the lower voltage. At 1600 volts the CsI crystal would have even greater pulse heights and a more extended plateau. Table IV and Fig. 1 show that CsI and also CsBr are excellent crystals for counting with alpha-particles and probably with other highly ionizing par-

ticles such as protons. With gamma-rays they have smaller peak heights than efficient organic crystals; nevertheless for the reasons mentioned above, especially CsI may be quite effective for counting with weak gamma-radiation. For counting NaCl is not good since its average peak height is small and can scarcely be distinguished from pulses of single photons which occur because of the large phosphorescence of these crystals. It is interesting to note that NaCl crystals activated with 5 percent AgCl have greater peak heights, though their ability to be stimulated by light after being exposed to high energy radiation is inferior to that of the 1 percent crystals.

The reasons for the difference between integrated intensities and peak heights and the relative gamma-ray and alpha-particle induced peak heights are associated with the different light emission times associated with these crystals. The peak heights were measured with a 1-megohm resistor at the output of the photomultiplier and since the capacity at the output is about 10 mmf,

TABLE IV. Relative peak voltages for various substances.<sup>a</sup>

Substance	Gamma-ray irradiation	Alpha-particle irradiation
Single crystals		
Anthracene	42	57
Stilbene	51 <sup>b</sup>	54
Naphthalene	17	31
Potassium iodide (Tl)	13	43
Calcium tungstate	19	43
Cadmium tungstate	11	24
Cesium bromide (Tl)	20	88
Potassium bromide (Tl)	7	280
Cesium iodide (Tl)	27	175
Sodium chloride (5% AgCl)	12	25
Sodium chloride (1% AgCl)	9	Noise
Powders		
ZnS (long persistent, Ag)	...	410
ZnS (short persistent, ZnCd)	...	340
ZnO	...	60
Solutions		
Phenylcyclohexane + <i>p</i> -terphenyl (3 g/l)	17	...
Xylene + <i>p</i> -terphenyl (5 g/l)	17	...
Xylene + <i>m</i> -terphenyl (15 g/l)	9	...
Xylene + <i>m</i> -terphenyl (200 g/l)	13	...
Phenylcyclohexane + <i>p</i> -terphenyl (3 g/l) + diphenylhexatriene (0.015 g/l)	21	...
Phenylcyclohexane + diphenylhexatriene (0.7 g/l)	14	...
Phenylcyclohexane + diphenylhexatriene (4 g/l)	11	...

<sup>a</sup> All measurements were made with 1700 volts at the 1P21 photomultiplier. All voltages are referred to *R* equal to 1 megohm.

<sup>b</sup> Note added in proof: Other investigators have usually found anthracene to have greater peak heights than stilbene. Our integrated measurements (Kallmann, Furst, and Sidran, *Nucleonics*, September, 1952) have also shown anthracene to be more efficient. The special crystals under investigation definitely showed greater peak heights with stilbene for gamma-rays.

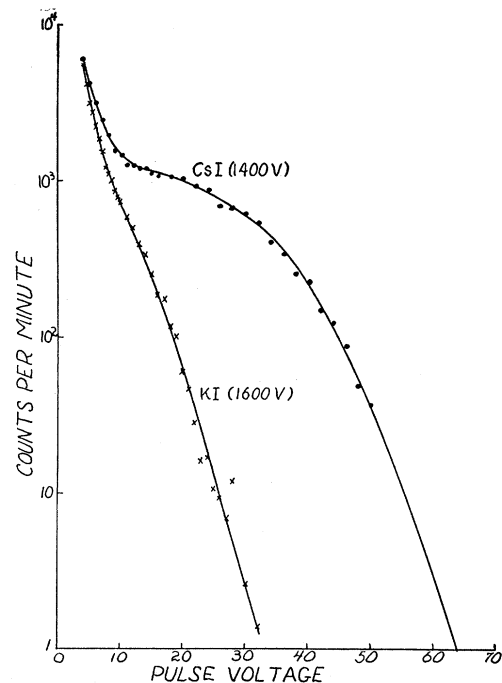


FIG. 1. Peak height distribution for cesium iodide and potassium iodide crystals activated with thallium.

the charging time of the capacity does not greatly exceed a period of the order of 10 microseconds. Only those light photons which strike the photomultiplier within this time interval make a contribution to the maximum peak height. If, however, the emission time is longer than 10  $\mu$ sec, only a certain fraction of the light flash will be utilized to charge the capacity to its maximum voltage. Thus all substances with longer emission times give smaller peaks, though the integrated intensities may be equal. Thus, the small peak heights of sodium chloride is an indication of an extremely long emission time; KBr also has a rather long emission period; cesium bromide and cesium iodide have shorter emission periods.

Preliminary measurements on the actual time constants bear out these indications of the magnitude of the time constant from comparison of the peak heights and integrated intensities. In the case of CsI and CsBr the emission time extends to more than 10  $\mu$ sec. However, they may still be valuable for counting heavy particles, especially at low counting rates. These crystals have peak heights smaller than those of the best available ZnS by a factor of 2; they have, however, the great advantage of being easily available in the form of large single transparent crystals.

We would like to acknowledge the help of Norman E. Linson in obtaining some of the data.