of nickel 61 or 62. Inasmuch as copper 66 exhibits an activity of 5 minutes, it was deemed advisable to free the nickel 64 isotope of any copper and zinc which might accidently be present. This chemical procedure was carefully done with the isotope appearing for bombardment as an oxide. In a subsequent neutron bombardment of the sample, the 4- to 5-minute activity persisted, suggesting that the activity is produced from a neutron bombardment of nickel 64. As an aid to the determination of this half-life, it was found useful to count the activity through sufficient absorber to eliminate the 1.75-hour half-life produced from the fraction of Ni<sup>61</sup> existing in this sample. This left the 2.6-hour half-life as a good base line from which to proceed in the analysis of the decay curve. The activities remaining were the 13.9-minute Co<sup>62</sup> from Ni<sup>62</sup> in this sample, and the 4- to 5-minute period. The latter has been detected through as much as  $1250 \text{ mg/cm}^2$  of aluminum.

Because of the shortness of the half-life involved and to the very small size of the sample of Ni<sup>64</sup>, it has been difficult to establish certainly whether the source of the activity is in the cobalt or nickel group. Present chemical evidence points to cobalt with the suggestion that we are dealing with an n,p reaction on nickel 64 producing cobalt 64.

## V. ACKNOWLEDGMENTS

The authors wish to acknowledge the cooperation of Messrs. Duane Sewell and James Vale of the 184-in. cyclotron staff, and Thomas Putnam and B. Rossi of the Crocker Laboratory 60-in. cyclotron group. Mr. Putnam contributed the use of special counting equipment for the study of short half-lives in the measurements of the 1.6-minute activity.

The metallic samples of isotopic nickel were prepared by J. Beaufait by an electroplating and stripping process.

The calutron analyses were carried out under the direction of Dr. Keith Pierce.

This problem was carried out under the auspices of the Atomic Energy Commission.

PHYSICAL REVIEW

VOLUME 75, NUMBER 4

FEBRUARY 15, 1949

## Quantitative Measurements with Scintillation Counters

HARTMUT KALLMANN Kaiser-Wilhelm-Institute for Physical Chemistry, Berlin-Dahlem, Germany (Received June 10, 1948)

The efficiency of a number of fluorescent materials used as scintillation counters for various materials has been measured. Two characteristics have been distinguished: (a) The physical light yield, i.e., the fraction of the absorbed energy transformed into light, and (b) the practical light yield, i.e., the amount of light obtained from a given intensity of radiation of a particular type, with the thickness of the phosphor adjusted for optimum results. Although the sulfide phosphors are high in physical yield, their practical yield is relatively lower because they are quite opaque to their fluorescent radiations. The organic phosphors, such as naphthalene and phenanthrene, and the potassium bromide phosphors are very much better in this respect.

THE counting of radioactive radiations by means of scintillations detected by photomultiplier tubes has recently received much interest.<sup>1</sup> We have performed a series of experiments with various fluorescent materials to investigate the applicability of this method to the determination of the intensity and energy of various radiations.<sup>2</sup> We characterize various phosphors by two main characteristics: (a) The physical light yield which is the fraction of the absorbed radiation energy transformed into

<sup>&</sup>lt;sup>1</sup> See, e.g., F. Marshall and J. W. Coltman, Phys. Rev. **72**, 528A (1947); R. J. Moon, Phys. Rev. **73**, 1210 (1948); P. R. Bell, Phys. Rev. **73**, 1405 (1948); G. B. Collins and R. C. Hoyt, Phys. Rev. **73**, 1259 (1948).

<sup>&</sup>lt;sup>2</sup> See also I. Broser and H. Kallmann, Zeits. f. Naturf. 2a, 439 (1947), Natur and Technik (July, 1947).

	Wave- length	Physical (relative to ZnS)							Technical (relative to ZnS)					Energy yield
		x-ray $\lambda$ (A)							x-ray λ (A)					
Phosphor	Ă	α	β	γ	1.93	0.63	0.28	α	ß	γ	1.93	0.63	0.28	ά
ZnS-Ag	4500	100	100	100	100	100	100	100	100	100	100	100	100	0.28
ZnS-Cu	5200	140	160	160			240	140		600			450	0.25
ZnS/CdS-Cu	5900	46						46						0.092
CdS-Ag	7600	78	$\sim 80$	$\sim 80$				78						0.23
Zn <sub>2</sub> SiO <sub>4</sub> -Mn	5250	13.5	81	80	40	50	52	13.5	130	130	40	50	60	0.027
ZnO	5500	7.0						7.0						0.014
CaWO4	4300	6.3	49	60	18.5	23	30	6.3	52	72	25	33	36	0.013
MgWO <sub>4</sub>	4900	6.4						6.4						0.013
KBr-Tl	3600	6	50.4	50.4				6		>1700				0.017
Naphthalene	3600	20	35	35	9	11		2.0	220	≥1000				0.004
Diphenyl	3600	6.0	52.5	525	25	22		6.0	220	580				0.012
Phenanthrene	4500	3.6	∞105	>61				3.6	198	241				0.007

TABLE I. Light yields of various phosphors.

light. (b) The technical light yield which gives the amount of light obtainable from a given intensity of radiation of particular type, choosing the optimum thickness of phosphor. The technical light yield depends on the physical light yield, the transparency of the phosphor for its own radiation, and the absorption for the primary radiation. It is desirable that a large fraction of the incident radiation be absorbed in a layer which is still transparent for the emitted light.

The physical light yield of the sulfide phosphors for  $\alpha$ -particles approaches the maximum possible. E.g., for zinc sulfide it was found that for  $\alpha$ -particles (independent of their energy) one light quantum is emitted for about 9 ev of kinetic energy expended in the phosphor, indicating a physical light yield of 27 percent.

It was found that with this phosphor an incident beam of 100  $\alpha$ -particles/sec. caused steady output current of about 10<sup>-9</sup> amp. This current can be easily measured particularly when the tube is cooled to reduce the dark current. The multiplier has a gain of about 10<sup>5</sup>, and the photo-cathode an efficiency of about 10<sup>-2</sup> electron/photon. The physical light yield of ZnS for

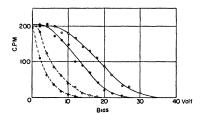


FIG. 1. Pulse height for Po  $\alpha$ -particles after their passage through various absorbers. ZnS phosphor.

irradiation by  $\beta$ -particles is lower,<sup>3</sup> perhaps by a factor of two. In the third column of Table I the physical light yield of a variety of phosphors under alpha-bombardment is tabulated. The values should be accurate to about 15 percent. Because of the much greater uncertainty in the determination of the physical light yield under electron bombardment, the values for all phosphors have been referred arbitrarily to a value of 100 for ZnS-Ag, for each radiation considered. All values have been recalculated for uniform spectral response and refer to energy yield, not quantum yield. These values are tabulated in columns 3-8 in Table I. All values refer to steady currents, not pulses. Although the physical light yield of ZnS phosphors approaches perfection, the technical yield presents a very different picture. Most conventional phosphors are quite opaque to their fluorescent radiations. Thus only the light produced in about 50 mg/cm<sup>2</sup> nearest the photo-tube window is effective. In

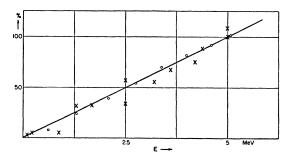


FIG. 2. Light output for  $\alpha$ -particles of different energies. Circles: single pulses; crosses: steady current. ZnS phosphor.

<sup>3</sup> H. J. Born, N. Riehl, and K. G. Zimmer, Physik. Reichsber. 1, 153 (1944).

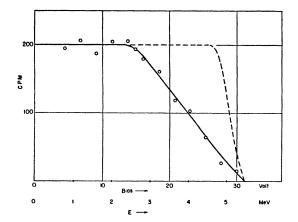


FIG. 3. Bias curve for Po  $\alpha$ -particles using a single crystal CdS phosphor.

the case of  $\alpha$ -particle bombardment this results in a drop in the output as the thickness is increased beyond about 50 mg/cm<sup>2</sup>. For more penetrating radiations, particularly gamma-rays, it results in a "saturation" at about this thickness. We have investigated two other groups of phosphors which are more transparent to their fluorescent radiation. These are: (a) Organic phosphors, particularly those containing conjugate double bonds like naphthalene and phenanthrene,<sup>4</sup> and (b) potassium bromide<sup>5</sup> phosphors. The organic phosphors are characterized by their extreme transparency; e.g., sublimed, carefully purified naphthalene is still perfectly transparent for its own fluourescent light, in layers of 1-cm thickness. Diphenyl and phenanthrene, which have a greater physical light yield than naphthalene, show some absorption which can, however, be eliminated by producing transparent blocks by solidification from a melt. These blocks are perfectly clear and transparent to their fluorescent light but have a somewhat lower physical light yield than when the material is produced by other methods.<sup>6</sup> The great transparency of these organic phosphors makes them particularly suitable for the counting of energetic electrons and gamma-rays. Another advantage of the organic phosphors for the counting of individual scintillations is the short duration of the light flash—less than 10<sup>-6</sup> second. In this respect

FIG. 4. Pulse height of uranium fission fragments (solid curve). Dotted line: uranium  $\alpha$ -particles.

they are much superior to the other phosphors. A more complete report on the investigations on the counting properties of the organic phosphors, particularly naphthalene, will appear soon in The Review of Scientific Instruments.

Professor Neuhaus (Marburg) has kindly prepared several large single crystals of potassium bromide phosphors. These also show a very high physical light yield for energetic electrons. However, the several large crystals showed varying light yields depending on the method of preparations. Our best crystal had an area of 3 cm<sup>2</sup> and depth of 1 cm, was perfectly transparent, and had a physical light yield of 0.5 compared with ZnS and 1.5 compared with naphthalene. Because of its relatively high density, it absorbs gamma-rays strongly, and its technical light vield for gamma-rays is therefore greatly superior even to the organic phosphors. An intensity of a few thousand gamma-quanta per second can still be detected by the steady current output of the photo-multiplier. Crystals of this general type may also prove very satisfactory as intensifying screens for the photographic recording of very hard x-rays. The particular crystal used by us emits light which is photographically not very effective since it is in the near ultraviolet. Other similar crystals can undoubtedly be prepared for greater photographic efficiency. On the other hand, the KBr phosphors are not suitable for the counting of single scintillation, since the duration of the flash is rather long according to our measurements.

Both the organic and the KBr phosphors are

<sup>&</sup>lt;sup>4</sup> More extensive reports in press in Zeits. f. Naturf. and Rev. Sci. Inst. <sup>5</sup> v. Meyeren, Zeits. f. Physik **61**, 321 (1930).

<sup>&</sup>lt;sup>6</sup> L. Herforth and H. Kallmann, Ann. d. Physik, in press.

less well suited for  $\alpha$ -particles, since their physical light yields are much lower under excitation by  $\alpha$ -particles.

Another possible application of these phosphors is to be conversion of electron beam images, e.g., in the electron microscope, as suggested by H. von Ardenne. For this purpose one wants not only transparent materials of considerable thickness to permit work with high energy electrons, but we should also prefer single crystals of sufficient size to avoid graininess of the screen. It is not certain whether the organic phosphors are suitable for operation in the vacuum of an electron microscope. The KBr phosphors would certainly be applicable and are available in large crystals. In the meantime we have also succeeded in preparing monocrystalline phosphors of ZnS and, particularly, CdS. The latter crystals were prepared by the methods of Frerichs<sup>7</sup> and then especially activated. ZnS crystals were prepared by W. Broser. CdS crystals can be prepared in thin pieces, several  $cm^2$  in area and about 100 mg/cm<sup>2</sup> thick. The fluorescence, on proper activation, is an intense red, near the limit of the visible region. The physical light yield is as good as that of ZnS powder. The duration of the light flash is rather long-of the order of a tenth millisecond. The main difficulty lies in the high refractive index of the materials which causes most of the light to escape from the narrow edges because of total internal reflexion. The edges must therefore be masked.

We have investigated the pulse height distribution of the multiplier output when thin layers of ZnS and single crystals of CdS are bombarded with  $\alpha$  particles. Figure 1 shows the (integral) bias curves for a ZnS screen for particles for various values of the residual range spent in the phosphors. Considerable pulse height straggling, which we believe to be due to the microcrystalline structure of the screen, appears in Fig. 2 the points marked by circles are obtained by integration of the curves in Fig. 1. These integrals should represent the average light output per particle of the particular residual range (and

therefore particular energy). The crosses were obtained by measuring the d.c. output of the multiplier for a strong sample of  $\alpha$ -particles. The fact that these points fall on a straight line indicates that the physical light yield is independent of particle energy. The pulse height straggling can be greatly reduced by the use of a monocrystalline CdS phosphor as shown in Fig. 3. The dotted line indicates the bias curve expected in the absence of instrumental straggling. The remaining spread of about  $\pm 20$ percent in pulse height can be still further reduced by masking out a small area of the phosphor.<sup>8</sup> One can clearly use this method to distinguish between particles of appreciably different energy down to 100 ev. We are using this method in our laboratory for the calibration of radioactive samples. This scintillation counter can also be used as a detector for range measurements. In this case the curves of Fig. 2 can be used to correct for counting losses near the end of the range. CdS gives consistently smaller pulses than ZnS, perhaps partly because of the greater duration of the pulses—about  $10^{-4}$  sec. in CdS compared with less than 10<sup>-5</sup> sec. in ZnS. Finally, we have investigated the pulses produced by fission fragments from a layer of uranium on the ZnS screen. Fissions were produced by a 10-mg Ra-Be source. Figure 4 shows the bias curve for the fission fragments. The fact that the pulses were really due to fission fragments was verified by the observation that a foil of  $1.5 \text{ mg/cm}^2$  between the uranium and the ZnS screen completely suppressed the large pulses. It is noteworthy that the  $\alpha$ -particle pulses from the uranium were completely unaffected by the gamma-rays from the Ra-Be source. Analogous experiments with gamma-rays, using naphthalene as phosphor, will be described in another publication.

Note added in proof: Since the writing of this paper we have developed better cadmium sulfide crystals giving curves which almost follow the dotted line of Fig. 3. These crystals give nearly homogeneous flashes for monoenergitic alphaparticles.

<sup>&</sup>lt;sup>7</sup> R. Frerichs, Naturwiss. **33**, 381 (1946); Phys. Rev. **72**, 594 (1947).

<sup>&</sup>lt;sup>8</sup> I. Broser and H. Kallmann, Ann. d. Phys. 3, 281 (1948).