Response of NaI(Tl), KI(Tl), and Stilbene to Fission Fragments

J. C. D. MILTON AND J. S. FRASER
Chalk River Laboratory, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada
(Received August 30, 1954)

The scintillations produced in NaI(Tl), KI(Tl), and stilbene by fission fragments are found to be about twice as large as those produced by 5-Mev alpha particles. The superior resolution of the inorganic crystals clearly separates the light and heavy fragment groups. The larger scintillations are associated with the more energetic light-fragment group. The pulse height as a function of residual range was measured for both groups in KI(Tl), whence it was found that at large values of the specific energy loss the specific fluorescence again increases rapidly.

I. INTRODUCTION

ALTHOUGH a great deal has been done on the scintillation response of both inorganic and organic phosphors to light particles[1] and light nuclei[2] such as H, D, He, and Ne, nothing is known about the response to energetic, heavy, multiply ionized particles such as fission fragments. Since most phosphors, particularly the organic ones, are showing signs of saturation[3] for α particles with energies of a few Mev, and since the mean range of the more energetic light fragment group is about $\frac{3}{4}$ that of a 5-Mev α particle, it might be thought that fission fragment scintillations would be smaller than those of α particles. However, in all 3 phosphors investigated, the fission fragment pulses were larger. Apart from its intrinsic interest, knowledge of the scintillation response is critical in all experiments involving fission fragments and fast coincidence techniques, where scintillation counters are desirable.

II. EXPERIMENTAL PROCEDURE

The apparatus used in comparing the crystals is shown in Fig. 1. It consists of a small vacuum chamber made of aluminum tubing (0.050-in. wall) with a 5819 photomultiplier inserted at one end. The source of fissionable material used was U^{233}, 180 $\mu g/cm^2$ thick, electroplated on a 1-in. diameter nickel disk placed in a beam of slow neutrons from the thermal column of the NRX reactor at the Chalk River Laboratory. The source was mounted on a long thin aluminum stand-off to reduce the amount of material in the beam. The crystals were mounted on a thin Lucite light pipe, about 3 mm thick at the center. All optical contacts were made by Dow-Corning "200" silicone fluid.

The NaI(Tl) and KI(Tl) crystals[4] were 1 cm on edge and $\frac{1}{2}$ mm thick. The NaI was cleaved and placed on the light pipe in a dry box through which moisture free argon was circulated. The vacuum chamber was then assembled, removed from the dry box and evacuated. The stilbene crystal,[5] 5 mg/cm^2 thick, $1\frac{1}{4}$ in. in diameter, was mounted on a light pipe of the same dimensions as the one used for the two inorganic crystals. The crystals were $1\frac{3}{4}$ in. away from the source.

The amplified pulses from the photomultiplier were analyzed by a 30-channel Marconi kicksorter. As there is a large variation in pulse height from one crystal to another, the pulse height scale was calibrated in the units of a standard pulser which fed pulses through a small condenser permanently attached to the input of the amplifier system. After crystals were changed,

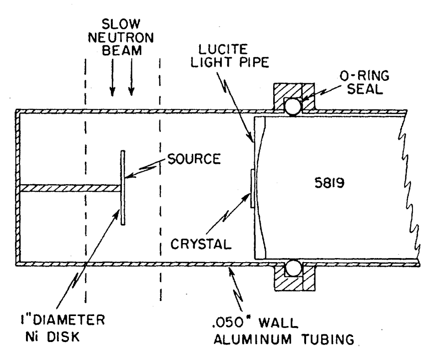

FIG. 1. Schematic drawing of vacuum chamber used in the comparison of phosphors.

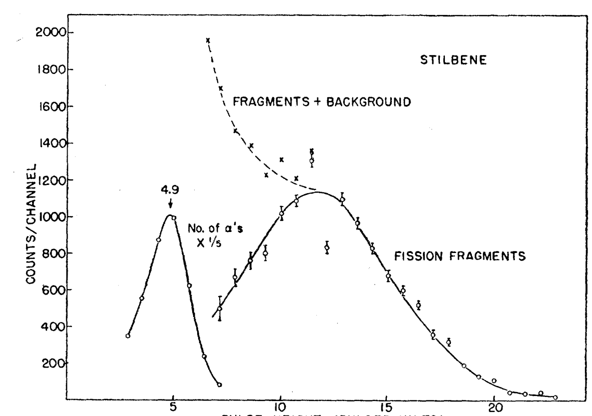

FIG. 2. Pulse height spectrum of fission fragments on stilbene.

[1] See, e.g., Taylor, Jentschke, Remley, Eby, and Kruger, Phys. Rev. 84, 1034 (1951).
[2] S. K. Allison and H. Casson, Phys. Rev. 90, 880 (1953).
[3] J. B. Birks, Phys. Rev. 84, 364 (1951); Phys. Rev. 86, 568 (1952).
[4] Supplied by Harshaw Chemical Company, Cleveland, Ohio.
[5] Supplied by Larco Nuclear Instrument Company, Palisades Park, New Jersey.

the voltage on the photomultiplier was returned to its previous value to within 0.1 percent. The data were taken in a single morning so that errors due to slow drifts were minimized.

III. RESULTS

The results are shown in Figs. 2–4. For comparison the 4.82-Mev U^{233} α particles are shown. The counting times for the fission fragments and α particles were not the same so that the relative number of counts is not significant. Only the position of the peak is important. The two fission fragment groups are well resolved by the inorganic crystals, but not by the stilbene. In all three cases, however, the fission fragment pulses are larger than the α pulses. The relative pulse heights are summarized in Table I. The relative α pulse heights agree well with the results of previous workers.[6]

If the width of the α peak in KI is assumed to be due solely to the statistical fluctuations of photoelectron production in the photomultiplier, then a width for the NaI α peak may be computed from the relative pulse heights. The observed width is greater than the calculated one. If then we use the fission fragment distribution observed in KI and from it compute the distribution in NaI, taking into account the poorer resolution necessary to give the observed α-peak width in NaI, we obtain a curve in good agreement with the experimentally observed fragment distribution. This suggests that the poorer fission fragment resolution of NaI is not something intrinsic, but is connected with the particular crystal used,—is perhaps caused by non-uniformity, or slight surface deterioration. Similarly in stilbene the observed α-peak width is again wider than the one computed from the KI results. In this case the thickness of the crystal is comparable to the range of the α particles so that thin spots in the crystal may contribute a low-energy tail. The width of the α peak is such that one would not expect to resolve the two fission-fragment groups.

TABLE I. Relative pulse heights.

	Light/α	Heavy/α	Alpha
NaI	2.1	1.6	17.8
KI	3.5	2.3	3.8
Stilbene	2		1

TABLE II. Fast coincidence figure of merit for three phosphors.

Phosphor	KI	NaI	Stilbene
$\bar{t}$(mμsec)	3	0.2	0.1

For fast-coincidence work a figure of merit may be defined as a number proportional to the average time $\bar{t}$ taken for the first photoelectron to leave the photocathode;[7] $\bar{t}=\tau/\bar{R}$, where τ is the mean life of the scintillations (the decay time) and $\bar{R}$ is the average number of photoelectrons emitted per pulse. The value of $\bar{t}$ depends on the photomultiplier and light collecting system used. The numbers shown in Table II are for the 5819 used in this experiment, an average tube. The values shown in the table were computed for the most probable light-fragment pulse height in the cases of NaI and KI, and for the most probable pulse height in the case of stilbene. Where the added energy resolution is important, use of NaI is indicated despite the slight decrease in speed.

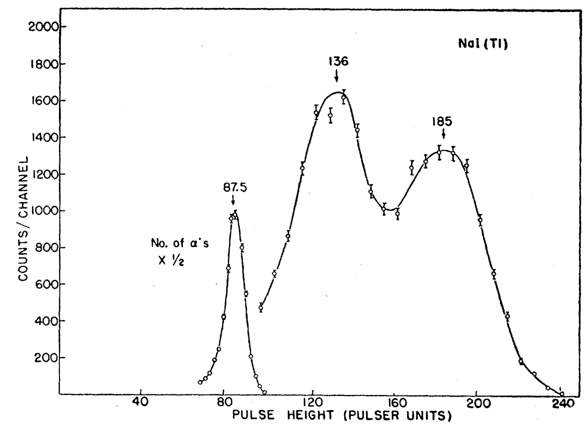

FIG. 3. Pulse height spectrum of fission fragments on NaI(Tl).

IV. FURTHER EXPERIMENTS WITH KI

In order to gain more information on the scintillation response to fission fragments, two further experiments with KI were performed. The first established that the larger scintillations were produced by the more energetic light-fragment group.

Identification of Fragment Group with Pulse Size

A thin source, a few μg of U^{233} in a collodion foil, was mounted in an ionization chamber filled to a pressure of

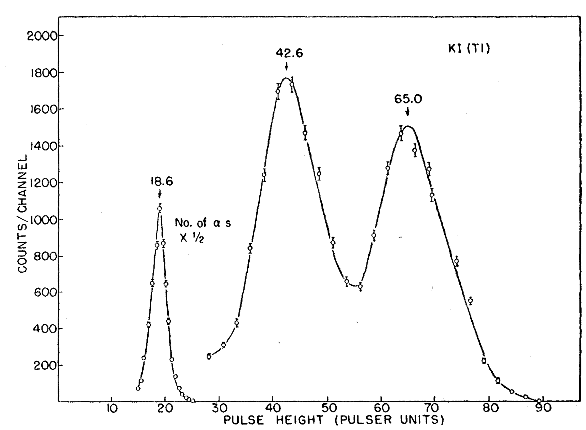

FIG. 4. Pulse height spectrum of fission fragments on KI(Tl).

[6] E.g., R. Hofstadter, Nucleonics 6, No. 5, p. 70 (1950).

[7] R. F. Post and L. I. Schiff, Phys. Rev. 80, 1113 (1950).

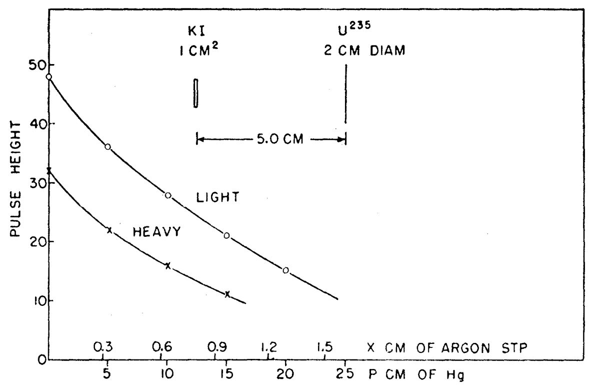

FIG. 5. Pulse height *versus* range for fission fragments in KI showing the source and crystal geometry used.

20 cm of Hg with argon plus CO_2. One fragment entered the ion chamber while the paired fragment entered a KI crystal 1 cm from the source. A single channel analyzer was set on one of the groups of pulses from the scintillation counter and the output used to gate the 30-channel kicksorter recording the ionization chamber pulses. In this way it was found that large scintillation pulses were paired with small ionization pulses and *vice versa.*

Specific Fluorescence of KI

The final experiment was designed to measure the specific fluorescence dL/dx as a function of the residual range of the fragment. To avoid confusion of the fragment pulses with α-particle pulses, the source used was of uranium enriched to 14 percent U^{235} and electroplated on a nickel disk to a thickness of 180 $\mu g/cm^2$. The arrangement of source and crystal is shown schematically in Fig. 5. By varying the pressure of argon in the chamber the fragments could be suitably

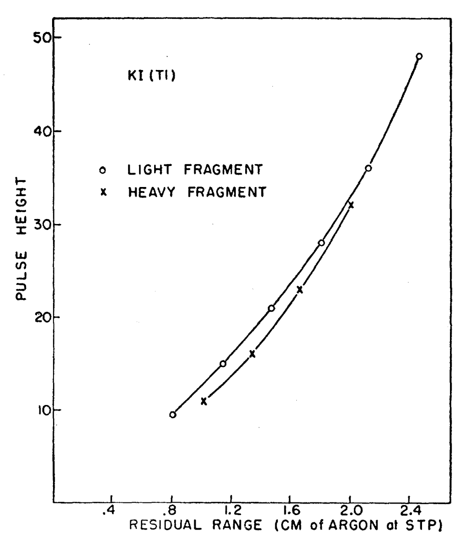

FIG. 6. Pulse height *versus* residual range for fission fragments in KI (see text).

slowed down before entering the crystal. The scintillation pulses were recorded on a 30-channel kicksorter. The peaks of the light- and heavy-fragment groups are plotted as a function of pressure in Fig. 5. Knowing the pressure and the distance, the equivalent distance travelled in argon at STP can be calculated and is also shown in Fig. 5.

From these data we calculated the scintillation response as a function of residual range and of energy using the range-energy relations given by Lassen.[8] The results are shown in Figs. 6 and 7. These curves should be considered only as illustrative of general trends because of the numerous uncertainties involved. For instance, the relative position of the light- and heavy-fragment points on the pulse height-range curve depends on the assumed maximum range. A shift of

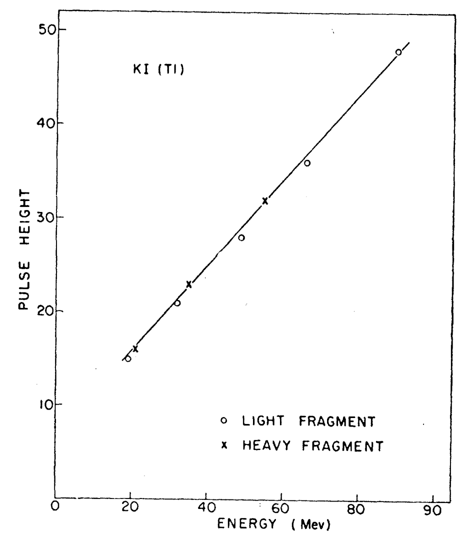

FIG. 7. Pulse height *versus* energy for fission fragments in KI (see text).

0.08 cm in the maximum range of one of the fragments would put the two curves into apparent coincidence. There is some doubt as to the correct range to use, and such a shift is entirely possible. A second instance is the apparent positive intercept on the pulse height axis of the extrapolated pulse height-energy curve. In this case the range-energy curves of Lassen were based on an energy of 86 Mev for the most probable light fragment energy whereas a better value now is 98.4 Mev.[9] We have not attempted to correct Lassen's data because of the unknown variation of the ionization defect[9] with energy.

The data can be treated in one further way, by plotting specific fluorescence dL/dx as a function of the specific energy loss dE/dx. This has been done in Fig. 8. In the derivation of the values of dE/dx, KI was

[8] N. O. Lassen, Kgl. Danske Videnskab. Selskabs, Mat.-fys. Medd. **25**, No. 11 (1949).
[9] R. B. Leachman, Phys. Rev. **87**, 444 (1952).

considered as equivalent to an equal, condensed mass of 50 percent argon plus 50 percent xenon so that Lassen's[8] results for these two gases could be used. The specific fluorescence was obtained by differentiating the curves of Fig. 6. The uncertainties shown in Fig. 8 are the estimated errors in determining the derivative. Despite these uncertainties there is definite evidence for an increase of the slope at the higher energy losses, but no real difference between the light and heavy fragments. The rapid increase of dL/dx at high dE/dx is quite different from the behavior of NaI under α bombardment at $dE/dx\sim 1$ Mev/cm as observed by Taylor *et al.*[1]

Such a curve may be fitted by a formula of the type suggested by Chou:[10]

$$\frac{dL}{dx}=\frac{A\,dE/dx}{a+b\,dE/dx+c(dE/dx)^2}, \tag{1}$$

for organic scintillators, but not by the simpler formula suggested earlier by Birks[11] in which $c=0$. The solid line shown in Fig. 8 has been computed from Eq. (1) by putting $a=0$, $b=0.0634$, $c=-4.73\times10^{-4}$. The zero value for a is not significant as the results in the region of large dE/dx are insensitive to this parameter. The previously published results for KI(Tl) in the region of small energy loss are contradictory: using α particles of energies up to 17 Mev, Franzen[12] found a linear relation between L and E; Link and Walker[13] (40-Mev maximum) obtained a plot of L *vs* E which was concave towards the energy axis, while Raffle and Robbins[14] (8.8-Mev maximum) and Almqvist[15] (7-Mev maximum) found one which was convex. If we assume, as found by Raffle and Robbins, and Almqvist, that the behavior of KI(Tl) is similar to that of NaI(Tl),[1,16] then b and a in Eq. (1) would be approximately equal. If a were made equal to 0.06, the resulting curve in Fig. 8 would be indistinguishable from that shown. Before any great validity is ascribed to Eq. (1), it would be necessary to have better data on the scintillation response to α particles up to 30 Mev and a more exact knowledge of the range-energy relation for fission fragments.

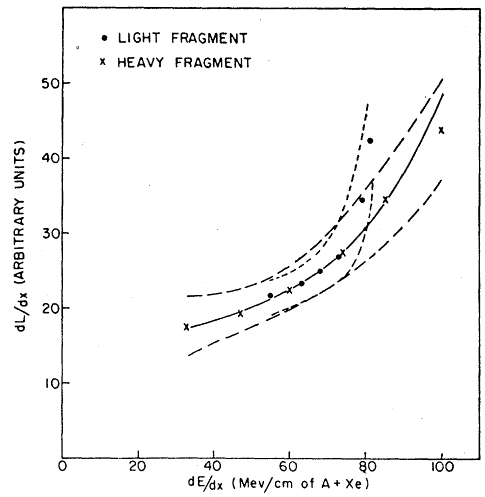

FIG. 8. Specific fluorescence as a function of the specific energy loss for fission fragments in KI (see text). dE/dx calculated from the data of Lassen assuming KI is equivalent to an equal mixture of argon plus xenon. The dotted and dashed lines are estimated limits of error in dL/dx. The solid line is computed from Eq. (1) with $a=0$, $b=0.063$, $c=-4.73\times10^{-4}$.

Some general conclusions may be derived from the curves of Figs. 5 and 8. It will be seen that most of the light is produced near the beginning of the fission fragment track where the rate of energy loss is high. This means, first of all, that there can be no large contribution to the light output made by the nuclear recoils[17,18] since these all occur near the end of the track where the fragment is moving slowly. Secondly, it means that surface effects will be extremely important. Thus, in a deliquescent substance such as NaI it is quite reasonable that the fission fragment pulses, relative to the alpha pulses, would be smaller than in the more stable KI.

[10] C. N. Chou, Phys. Rev. **87**, 904 (1952).
[11] J. B. Birks, Phys. Rev. **84**, 364 (1951); *Scintillation Counters* (Pergamon Press, London, 1953).
[12] Franzen, Peelle, and Sherr, Phys. Rev. **79**, 742 (1950).
[13] W. T. Link and D. Walker, Proc. Phys. Soc. (London) **A66**, 767 (1953).
[14] J. E. Raffle and E. J. Robbins, Proc. Phys. Soc. (London) **B65**, 320 (1952).
[15] E. Almqvist, Thesis, University of Liverpool, 1954 (unpublished).
[16] R. H. Lovberg, Phys. Rev. **84**, 852 (1952).
[17] Boggild, Brostrom, and Lauritsen, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **18**, No. 4 (1940).
[18] Niels Bohr, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **18**, No. 8 (1948).