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GAMMA RAYS ASSOCIATED WITH FISSION

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I. INTRODUCTION

Although many properties of the fission fragments and fast neutrons from U-235 fission have been measured in detail (1), corresponding studies for fission-associated gamma radiation have not been reported. This paper describes experiments designed to determine as a function of time after fission the absolute energy spectra of the gamma radiation emitted with the thermal fission of U-235**.

The energy release in the form of gamma radiation from U-235 fission, (13.1 ± 1.3) Mev per fission, represents a significant fraction (about 6%) of the total energy release per fission. Moreover, of the fission radiations, only neutrons compete with gamma rays for the position of greatest importance to the design of nuclear reactor shields (2). In order to design an efficient reactor shield, the absolute energy spectrum of the emitted gamma radiation must be known, since the cross sections pertinent to the attenuation processes are strongly energy dependent. For a shut-down reactor or for used reactor fuel, the shielding requirements are determined almost entirely by the presence of gamma rays emitted by the radioactive fission products.

An additional reactor-design problem is the removal of heat produced by gamma-ray interactions both in the reactor core and in the surrounding regions. Special cooling systems must frequently be provided for reflector, pressure shell, and shield regions of a reactor, and for experimental apparatus used with research reactors. Efficient design of these cooling systems is dependent upon a knowledge of the energy spectra of the sources of gamma radiation associated with fission (3).

Predictions of the fission-associated gamma-ray spectra have not been attempted on the basis of the theories of fission. Therefore, experiments provide the only access to these spectra. Experiments to determine the gamma-ray spectrum in time coincidence with fission used for a source the U-235

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** Throughout this paper, references to fission imply fission of U-235 by thermal neutron capture.

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contained in an ionization chamber. For studies of the gamma rays following beta decay of the radioactive fission fragments, samples of U-235 were irradiated briefly and then transported to a gamma-ray spectrometer.

In this report, time periods after fission are defined as follows: the interval less than 5×10^{-8} sec after fission is "prompt", delay intervals from 5×10^{-8} to 10^{-5} sec are "short", and those from 1 to 10^4 sec are "long".* Gamma-ray spectra for the prompt-time region compare well with the results of previous experiments (1). The observation of gamma radiation in the short-delay time region with an integral photon intensity 6% of the prompt radiation was unexpected. The spectral data for the long-time region describes the emission of approximately 75% of the gamma-ray energy delayed by times longer than 1 sec.

In Sect. II of this report is presented a brief description of the apparatus. Details are given elsewhere (4). The observed energy spectra are presented in Sect. III, arranged in order of increasing times after fission. Section IV gives the results of time-distribution measurements for selected energy groups, again in order of increasing times. Conclusions are in Sect. V.

II. METHOD

A. Gamma-Ray Energy Spectrometer

The gamma-ray energy spectrometer was of the multiple-crystal scintillation type (5). One NaI(Tl) scintillation crystal (the "center" crystal) absorbed the energy of electrons produced by gamma radiation incident upon it. Auxiliary crystals, largely shielded from the source, detected secondary gamma rays from either the Compton or pair interaction processes in the "center" crystal. The signals from photomultipliers which detected the scintillations in each crystal were caused after amplification and shaping to operate a coincidence circuit. The output of this circuit allowed analysis of the height of the pulse from the center crystal when the corresponding gamma-ray interaction was positively identified. The analyzed pulses are proportional in height to the electron energy absorbed in the center crystal. If one auxiliary crystal detects a large-angle Compton-scattered gamma ray, the "Compton spectrometer" is utilized. If both auxiliary crystals detect a gamma ray from the annihilation of a positron in the center crystal, the "pair spectrometer" is utilized. The former is useful for gamma rays of energy < 2.5 Mev, the latter for those of energy > 1.5 Mev.

Figures 1a and 1b show typical response curves obtained with the multiple-crystal spectrometers when radioactive sources were placed in the location normally occupied by the source of fission-associated radiation. These curves

* Although the term "long" is used, this data covers shorter decay periods than any comparable spectra. It is also important to distinguish between "time after fission" as shown here and "time after shutdown" of a fission source present in a neutron flux for an appreciable period.

Fig. 1 - Typical response characteristics to incident monoenergetic gamma rays for a) the two-crystal Compton spectrometer, and b) the three-crystal pair spectrometer. Note that the absorbed energy is less than the incident energy labeled on each curve, so that on a scale of incident energy the peaks would be relatively more narrow. Typical data points are plotted where possible.

are typical for monoenergetic radiation incident on an energy spectrometer, in the sense that only a fraction of the total response occurs at a position on the pulse-height scale which corresponds to the initial energy. Other, unwanted, responses occur at positions which usually correspond to lower radiation energies. For a small single-crystal scintillation spectrometer, only a small fraction of the response to a high-energy gamma ray (above ~ 1 Mev) is at the pulse height corresponding to that energy. The response of the multiple-crystal spectrometer employed here is more concentrated than that of a single crystal because the nature of the gamma-ray interaction in the "center" crystal is specified. Values of the fractional response occurring under the peak exceeded 0.55 for all energies studied.

The measured response functions of the spectrometer were used to construct a response matrix designed to remove the undesired counts. A row of this matrix approximated the response function of the spectrometer for a given gamma-ray energy. Measured pulse-height spectra were converted to photon energy spectra by multiplication by the inverse of the response matrix.

The efficiency of the spectrometer was determined by measuring its response to gamma-ray sources for which the disintegration rates were known from measurements in a 4π ionization chamber standardized by beta-gamma coincidence counting. Interpolations between the few available points at high energies were made on the basis of the known iodine cross sections (6) after correction for self-absorption effects in the spectrometer.

Analysis of the data from the spectrometer required correction for backgrounds and for losses caused by high counting rates. These corrections were determined experimentally.

B. Time Analyzers

For time intervals in the "short" region, a linear voltage sweep was started when a fission was announced by a pulse from the fission chamber (7). Upon arrival of a pulse from the gamma-ray spectrometer, the sweep was stopped to form a pulse whose height was proportional to the elapsed time. The minimum time interval that could be examined by this method was 3×10^{-9} sec per channel, but statistical fluctuations in the timing signals limited timing accuracy to about 2×10^{-8} sec.

For time intervals in the "long" region, an exponential sweep was started by the removal of a uranium sample from the region of high neutron flux. This sweep acted upon the bias of a multichannel pulse-height analyzer, while all detected gamma rays within a pre-selected energy range gave rise to pulses of uniform height at the analyzer input. Thus a large number of pulses could be related to the time-of-fission using a single sample of U-235.

III. ENERGY SPECTRA OF FISSION-ASSOCIATED GAMMA RADIATIONS

A. Prompt Times

For the observation of the prompt-fission gamma-ray energy spectrum, that portion of the output of the gamma-ray spectrometer was observed which was coincident in time, within 5×10^{-8} sec, with the pulses from a fission chamber. Fissions were induced in 93% U-235 in a 1-cm dia. spiral-wound ionization chamber (8) by neutrons from the thermal column of the Bulk Shielding Facility at the Oak Ridge National Laboratory. The shielding surrounding the spectrometer and fission chamber was selected after a long series of shielding optimization studies (9). Lithium was used to prevent interactions of nearly thermal neutrons with the sodium-iodide crystals, and lead in thicknesses up to 10-in. was used to eliminate unwanted gamma radiation.

It was not possible to eliminate the detection of prompt-fission neutrons by the time-coincidence requirement or by the use of shielding. The interaction of these neutrons caused an error in the previous measurements (10) of as much as 20%, in the energy region from 0.05 Mev to 0.6 Mev. The extent of this background in the equipment used for the present experiments was determined by two methods. Attenuation measurements using polyethylene and lead were made for the radiation in coincidence with fission, and time-of-flight measurements over flight paths of 20 to 50 cm were made by the methods described in Sect. IV.

In order to reduce the extent of the prompt-neutron background the size of the center crystal was reduced. After revision, both of the methods indicated that the integral background due to prompt neutrons was from 0-4% in the energy range < 2 Mev (9). The pair-spectrometer response to such neutron-induced backgrounds is much less.

The energy spectrum of the prompt-fission gamma rays is shown in Fig. 2. Corrections for backgrounds, non-uniqueness of response, and efficiency have been made. The response correction was only approximate since a final analysis awaits the accumulation of more data. The total energy emitted promptly in the form of gamma rays is (7.2 ± 0.8) Mev per fission in the interval from 0.3 Mev to 10 Mev. The total number of photons emitted is (7.4 ± 0.8) per fission within the same energy region. The observed photon spectrum cannot be fit, even within a factor of two, by a negative exponential function of the gamma-ray energy.

Fig. 2 - The energy spectrum of gamma rays observed within 5×10^{-8} sec of fission. The ordinate errors shown were obtained from counting statistics, and the energy errors represent in each case the energy interval over which the results were averaged. The line is drawn only to connect the points. This plot represents a preliminary analysis of the data, and systematic errors as large as 15% may occur in some energy regions.

B. Short Times

Measurements were carried out in the short-delay time region although no fission gamma-ray emitters had previously been reported to have such short decay times (11). Because of the low intensities of the subject gamma radiation, a 2.5-cm single-crystal gamma-ray detector was employed for these measurements. Therefore pulse-height spectra rather than energy spectra are reported. Fissions were produced in a 0.5-cm-dia. spiral fission chamber by a neutron beam from the Graphite Reactor of the Oak Ridge National Laboratory. Gamma rays were recorded if they produced pulses in delayed coincidence with pulses from the fission chamber. With this arrangement no special discrimination was available against delayed neutrons from fission. However, changing the material adjacent to the detectors from lead to lithiated paraffin demonstrated that slowing down of prompt-fission neutrons was not responsible for the observed delayed radiation.

The observed pulse-height spectra are shown in Fig. 3 for times after fission ranging from 0.12×10^{-6} to 1.4×10^{-6} sec. Prominent peaks may be seen corresponding to gamma-ray energies of 0.19, 0.30, and 1.3 Mev. To demonstrate the reality of these gamma rays, similar measurements were made to search for radiations emitted before fission. None were observed.

Fig. 3 - Pulse-height spectra observed at short times after fission. The uppermost curve labeled "0" shows the pulse-height spectrum of the prompt radiation as obtained with the same detector geometry. The plotted points for this curve were obtained by multiplying by 10^7 the observed (counts) (fission) $^{-1}$ (Mev) $^{-1}$. Random backgrounds have been subtracted, statistical errors are shown, and the lines are fitted to the points to connect associated data.

An experiment, with two gamma-ray scintillators connected in delayed coincidence with the fission chamber, demonstrated that the 0.19 and 1.3 Mev gamma rays are not emitted in cascade with the 0.3 Mev gamma rays.

C. Long Times

For measurements of the gamma radiations emitted from the longer-lived radioactive fission products, the gamma-ray spectrometer was located adjacent to the Graphite Reactor. After irradiation for time intervals sufficiently short to be considered instantaneous, samples of 93% U-235 were transported in about 0.25 sec from the interior of the reactor to the spectrometer by a pneumatic apparatus. After pre-set waiting times, spectra were recorded for appropriate short counting times. Tens or hundreds of such samples were used to obtain each recorded spectrum. In a second arrangement, a continuous belt containing U-235 was moved in about 1.2 sec

Fig. 4. - Gamma-ray energy spectra observed "long" times after fission. Statistical errors are shown, and the lines were fitted to the points by eye. All data, except that for the uppermost curve in (b), were obtained at the stated times after fission. The uppermost curve in (b) is the spectrum emitted from the continuous fuel belt about 8×10^3 sec after irradiation of the belt commenced. (Use right-hand scale.) No response corrections were included.

to the gamma-ray spectrometer from a region of high neutron flux at the Lid Tank Shielding Facility of the Oak Ridge National Laboratory. Spectra from the first arrangement are associated with a fixed time after the fission event, while the continuously moving belt yields a spectrum corresponding to a steady-state between formation and decay of each fission product. Since in the latter experiment the requirement of coincidence with a fission event could not reduce the backgrounds, massive shielding was required surrounding the spectrometer. By using a combination of shielding materials effective in attenuating neutrons and gamma rays, the backgrounds were reduced to a negligible level in the first arrangement, and to a tractable level in the second.

The number of fissions in each experiment was determined by calculations based on the observed activation of gold foils in the appropriate neutron flux. The activation of these foils was determined by using a 4π ionization chamber. Corrections were made to compensate for depression of the neutron flux in the uranium samples.

The energy spectra obtained in the above experiments are exhibited in Fig. 4. The peaks in the energy spectra appear real. Analysis of their isotopic origins must await high-speed chemical separations of the fission products.(12)

IV. TIME DISTRIBUTIONS OF FISSION-ASSOCIATED GAMMA RADIATION

A. Short Times

The time analyzer described in Sect. II was used to determine the dependence upon time after fission of the gamma rays associated with fission. The time interval was measured between the occurrence of a fission event and the detection of a gamma ray within an energy range selected by a single-crystal pulse-height spectrometer. The decay curves, which are shown in Fig. 5 for various energy intervals, have been corrected for all backgrounds

Fig. 5 - Time dependence of the emission of gamma rays for times after fission less than 10^{-6} sec. The data are plotted in terms of $(\text{counts}) (\text{fission})^{-1} (\text{sec})^{-1}$, since a single-crystal sodium-iodide detector was used and thus the true energy spectrum was not determined. Data have been combined for adjacent channels on the time analyzer. Before combination, the time-channel width for most of the runs was $(9.3 \pm 0.2) \times 10^{-8}$ sec. The different symbols for a given energy group correspond to independent runs. Statistical errors are shown, and the lines are drawn to connect the related points.

and for the contribution caused by prompt gamma radiation. The latter was determined by examining the time region before fission. The measurements were extended to 10^{-5} sec after fission, but for times greater than 10^{-6} sec the observed rates did not differ from the random backgrounds.

As observed with the single-crystal spectrometer, the magnitude of the integral photon intensity delayed into the short region is $(5.7 \pm 0.3)\%$ relative to the prompt radiation. The variation of this delay fraction with gamma-ray energy is shown in Table I. The relative contribution of the long-time delayed radiation (Sect. IV. B) in this short-time region is $< 10^{-4}$.

TABLE I. FISSION GAMMA-RAY DELAY FRACTION AS A FUNCTION OF ENERGY

Average Gamma-Ray Energy (Mev)	Gamma-Ray Energy Interval (Mev)	The Ratio of the Number of Counts Observed in the Specified Energy Interval for Short-Delay Times Relative to the Number of Counts for Prompt Times
0.19	0.15 - 0.22	$0.20 \pm .04$
0.30	0.25 - 0.35	$0.07 \pm .03$
0.70	0.48 - 0.92	$0.038 \pm .01$
1.3	1.18 - 1.42	$0.020 \pm .004$
-	0.16 - 1.93	$0.057 \pm .003$

Fig. 6 - Time dependence of the emission of gamma rays, for long times. On the lower portion of the graph are shown the photon-intensity time distributions for six energy intervals. At the top of the graph is shown the observed integrated energy release and the compilation of Perkins (13) based on spectral studies of chemically separated fission products. (Use right-hand scale.) No correction for the spectrometer response function was made for the photon data and only a very approximate correction was made for the energy release curve.

B. Long Times

For decay studies in the long-time region, clean samples of U-235 were exposed in the interior of the Graphite Reactor and then automatically removed to the spectrometer. The time between removal of the sample and the detection of each gamma ray was determined by the exponential-sweep time analyzer. For these measurements the multiple-crystal spectrometer was used, therefore the quoted energy intervals correspond to actual gamma-ray energies.

The resulting time distributions are shown in Fig. 6 for each of several energy intervals. On the same graph are shown the results of an analysis of gamma-ray data available from spectral studies of chemically separated fission products (13). The data for fission-product gamma rays, as summarized in Table II, indicates an emission of (5.7 ± 0.7) Mev per fission for times after fission from 1 sec to 10^3 sec and for energies above 0.3 Mev.

TABLE II. FISSION PRODUCT GAMMA-RAY ENERGY EMISSION PER FISSION

Energy Range (Mev)	Time Range (sec)		Energy Emission (Mev/fission)	Integrated From:
	From:	To:		
.28 - 5.0	1.0	$- 1.8 \times 10^3$	4.4 ± 0.8	Fig. 6
.36 - 5.0	1.5	$- 8.0 \times 10^3$	4.1 ± 1.3	Fig. 4
.1 up	1.8×10^3	$- 1 \times 10^8$	$1.6 \pm ?$	Perkins (13)
.1 up	8.0×10^3	$- 1 \times 10^8$	$1.1 \pm ?$	Perkins (13)
.28 - 5.0	1	$- 1 \times 10^8$	5.9 ± 0.8	(Combination)
.36 - 5.0	1.5	$- 1 \times 10^8$	5.2 ± 1.3	(of Above)

V. CONCLUSIONS

The energy released in the form of prompt gamma rays, (7.2 ± 0.8) Mev, agrees with that reported on the basis of previous spectral measurements (10). The distribution in energy of the prompt-fission gamma rays appears to be continuous from the lowest energy measured, 0.3 Mev, to an apparent upper limit of about 8 Mev. Nuclear excitations of greater than this energy would usually lead to the emission of a neutron. The apparent peaks at higher energies will be studied further by improving the statistical accuracy of the data. It is expected that the observed continuous spectrum is caused by the decay of a statistical ensemble of many levels. In this connection, it may be noted that the corresponding prompt gamma-ray spectrum from Cf-252 spontaneous fission is similar to that shown in Fig. 2 (14).

Of the various theories of fission (15) none predict the gamma-ray spectral distributions. The evaporation model of Leachman (1) leads to an expected energy of about 4.0 Mev for emission in the form of de-excitation radiation, rather less than that observed.

The observance of gamma radiation in the short-time region with integral photon intensities as great as $(5.7 \pm 0.3)\%$ of the prompt radiation was unexpected. Since nuclear beta decay is energetically forbidden for decay times as short as 10^{-3} sec, these measured gamma rays must be assumed to arise from isomeric transitions. The energy distributions, which show peaks at energies of 0.19, 0.30, and 1.3 Mev, are consistent with the time distributions.

The total known energy release per fission for times after fission greater than 1 sec is (5.7 ± 0.7) Mev for energies above 0.3 Mev. For the longest time intervals measured here, comparison with chemical data (13) indicates close agreement in absolute magnitude for the energy released per fission. For most of the time region covered, no other spectra are available from either experiment or theory.

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Abstract

This paper describes measurements of the energy spectra and time distribution of the fission-associated gamma rays. Such gamma rays arise from the fission process or from the de-excitation of the fission-product nuclei and their daughters. The energy and time distributions are required for the correct design of shield and heat-disposition systems for power reactors. Further, an understanding of the fission process necessitates correlation of these distributions with theoretical predictions.

ENERGY SPECTRA. Preliminary energy spectra for the prompt gamma radiation from fission were reported by Leachman at the 1955 International Conference. New data for thermal neutron-induced fission in U-235 is presented here covering the energy range between 0.3 and 8.0 Mev. Gamma-ray energy analysis was carried out with a multiple-crystal scintillation spectrometer employing either Compton scattering or pair production to give "unique" pulse-height response. The importance of unique spectrometer response is explained. The gamma-ray spectrometer was operated in coincidence with a spiral-wound fission chamber by the use of a four-fold coincidence circuit having a resolving time of 6×10^{-8} sec. The undesired contribution to the count rate caused by the fast neutrons emitted in fission was reduced to 4% of the total. By contrast, the preliminary measurements referred to above included a neutron-induced background of approximately 20%.

mineral turnover in the flat and in the long bones which can be reasonably related to the differential growth properties of the individual bones. It is apparent, however, that a gross similarity exists between the separate bones and the entire skeleton.

A remarkable similarity was found when the Sr^{90} -turnover equations for the rat and the monkey were compared, and this similarity suggests that the turnover equation for the monkey may be a reasonable estimation of turnover of Sr^{90} in human bones. One may then estimate the fractions of adult human bone that equilibrate with Sr^{90} and the rate at which such equilibration occurs.

These are:

- 0.06% of bone turnover with a half time of 3 days
- 0.2% of bone turnover with a half time of 6 days
- 1.3% of bone turnover with a half time of 50 days
- 98% of bone turnover with a half time of 970 days

The average cumulative radiation in bone following acute exposure to Sr^{90} is largely determined in that portion of the skeleton in which turnover rate is slowest; in this case 98% of the bone accumulates 38% of the administered Sr^{90} and subsequently eliminates it with a half time of 970 days. It is noted that for man an average Sr^{90} turnover in bone of about 1000 days would give a reasonable agreement with the Sr^{90} content of human bone as reported by Libby and others over the period from 1953 to 1957.³

A human being exposed initially to a bone burden of 1 m μ C of Sr^{90} (assuming uniform distribution in a bone mass of 7000 g) would have the following retention of Sr^{90} and radiation exposure (if he were not subsequently re-exposed to Sr^{90}).

Time (yr)	Sr^{90} (m μ C)	Dose (mr/yr)	Accumulated exposure (mr)
0 (initial exposure)	1	2.8	0
2.7	0.5	2.0	5.4
5.4	0.25	1.0	8.1
8.1	0.13	0.5	9.5
∞	0	0	10.8

³W. F. Libby, Current Research Findings on Radioactive Fallout, Remarks delivered before the American Association for the Advancement of Science, Washington, D. C., October 12, 1956.