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GAMMA-RAY FALLOUT SPECTRA FOR OPERATION PLUMBBOB

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[redacted]earch and Development Technical Report USNRDL-TR-286

NS 081-001

8 December 1958

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by

J. M. Ferguson
W. E. Thompson
F. M. Tomnovec

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U.S. NAVAL RADIOLOGICAL DEFENSE LABORATORY

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GAMMA-RAY FALLOUT SPECTRA FOR OPERATION PLUMBBOB

Research and Development Technical Report USNRDL-TR-286

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Effects of Atomic Weapons

Technical Objective
AW-7

Nuclear Radiation Branch
C.S. Cook, Head

Nucleonics Division
A. Guthrie, Head

Scientific Director
P.C. Tompkins

Commanding Officer and Director
Captain J.H. McQuilkin, USN

[redacted]

U.S. NAVAL RADIOLOGICAL DEFENSE LABORATORY
San Francisco 24, California

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ABSTRACT

Gamma radiation from fallout particles from Shot Diablo and Shot Shasta of Operation PLUMBBOB has been analyzed at various times after Shot Diablo and once after Shot Shasta using a NaI(Tl) crystal spectrometer. The earliest analysis was made 96 minutes after Shot Diablo to provide information about early-time spectral characteristics of fallout radiation. Short-lived Sr^{92} was found to be an important contributor to the activity of the fallout at early times.

SUMMARY

Because of the paucity of data on gamma-ray spectra of fallout samples only a few hours old, the present analysis was undertaken. Spectra were taken at various times ranging from 96 to 3,964 minutes after Shot Diablo and one spectrum was taken 3,120 minutes after Shot Shasta.

The results, giving the relative intensities of each detectable gamma-ray line as a function of time after shot, are shown in Tables I and II.

ADMINISTRATIVE INFORMATION

Background of Work

This Laboratory has been prosecuting, on a continuing basis, an investigative program, "Nuclear Radiation Characteristics" sponsored by the Bureau of Ships. The primary objective of this program is to determine the characteristics of nuclear radiations associated with nuclear detonations, including instantaneous radiation and radiations from induced activities and from fallout. No satisfactory measurement of the early-time gamma-ray spectral characteristics of fallout had been made prior to Operation PLUMBBOB. At that operation, the Project 2.2 personnel had the opportunity to make measurements of such radiation beginning at 1-1/2 hours after shot time. This was done in the time interval between shots of primary interest to Project 2.2 using the same equipment used for the primary project work.

Authorization and Funding

This work was authorized by the Bureau of Ships, Technical Objective AW-7, Project Index No. NS 081-001, "Effects of Atomic Warheads and Radiological Shielding", based upon the Laboratory's Technical Program 8, problem 1 for FY 1958 entitled, "Measurement of Spectral Characteristics of Nuclear Radiation" and Program A-2, Problem 8 for FY 1959 entitled, "Neutron Radiations from Nuclear Weapons." Funds were authorized by the Bureau of Ships on allotment 178/58 budget project 30 for FY 1958 and allotment 178/59 budget project 30 for FY 1959.

Description of Work

The pulse-height distributions obtained from the early time gamma-ray fallout spectral measurements were kept after Operation PLUMBBOB until an appropriate amount of time could be devoted to analysis of the work. This analysis has now been completed and the results are given in this report.

REPORT OF INVESTIGATION

INTRODUCTION

This report deals with a brief investigation of the gamma-ray energy spectrum of fallout pellets. These pellets were recovered from Shot Diablo and Shot Shasta of Operation PLUMBBOB. The objective of the investigation was to determine the relative gamma-ray intensity as a function both of energy and time, and to determine which nuclides contribute importantly to the activity. Results were obtained for gamma rays ranging from 0 to 3 Mev, and for times ranging from 90 minutes to 2 days after the detonation.

PREVIOUS WORK

Very little data are available on the gamma-ray spectra of fallout samples which are only a few hours old. Gamma-ray spectra from fallout samples obtained at Operation REDWING have been reported by Triffet and LaRiviere[1] and by W.E. Thompson.[2] Those results are for large weapons and for times after detonation ranging from 51 hours to 1,558 hours. Some gamma-ray spectral data were also obtained at Operation TEAPOT.[3] One sample was measured as early as 9 hours after detonation but the spectra from no other samples were observed earlier than 20 hours after detonation. Hence they are not strictly comparable to the results presented here.

Some work has been done on the activity of reactor produced fission products. Although these investigations do not take fractionation and other processes peculiar to fallout into account, they do serve as useful guides for analysis of the data presented here. An experimental investigation of the gamma-ray spectrum of U^{235} thermal neutron fission products was conducted by D. H. Peirson.[4] He gives gamma-ray relative intensities for 1 to 10 days after bombardment, and identifies the nuclide emitting each of the principal gamma rays.

Bolles and Ballou[5] have computed the relative activity of each U^{235} fission product as a function of time. These computations are based on the theory of R.D. Present.[6]

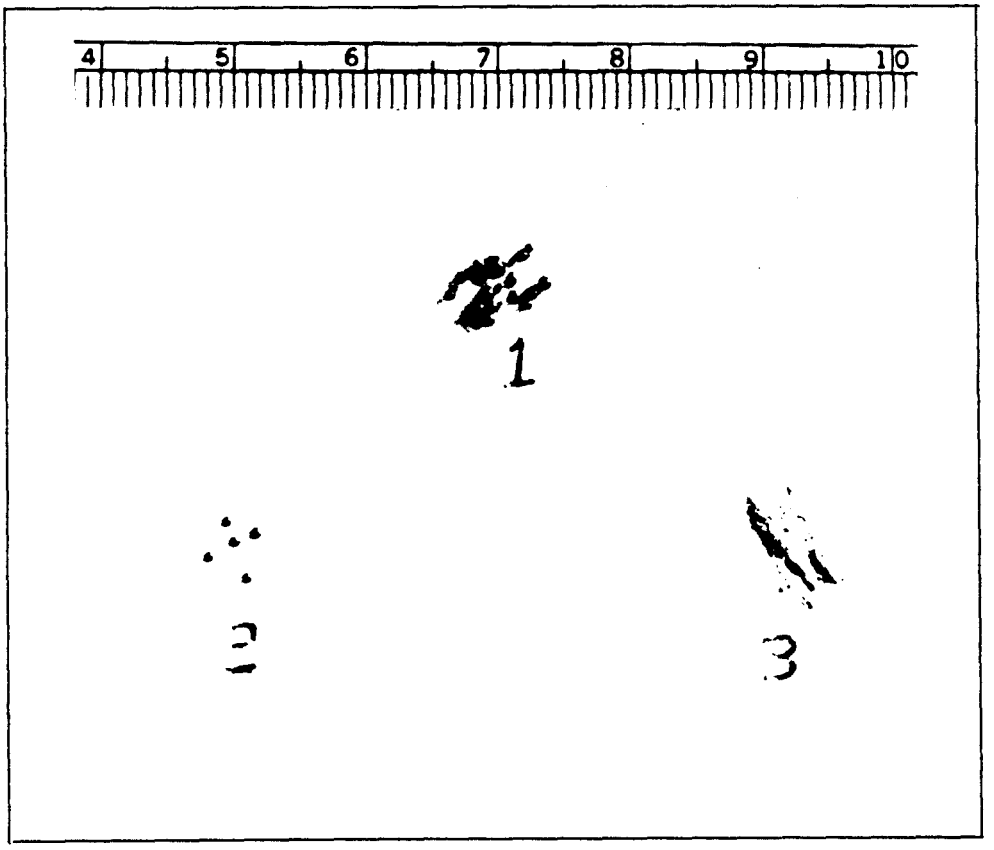

Fig. 1 Fallout Samples. Sample 1 consists of the pellets picked up by the magnet on the first scan. Sample 2 consists of those picked up on the second scan. Sample 3 is the residue left in the tray.

Blomeke[7] has compiled the nuclear properties of U^{235} fission products, including yield (measured or estimated), half period, and energy of beta and gamma radiation. Since gamma-ray energy assignments are based on a 1953 compilation,[8] many of these assignments are out of date.

NATURE OF THE SAMPLES

The fallout samples studied in this investigation were recovered from the Diablo and Shasta shots of Operation PLUMBBOB. Diablo was a 19 KT device and Shasta a 17 KT device. Both were detonated on 500-foot steel towers.

The open-closed fallout collectors[9] from which the samples were obtained were placed 5,300 feet from ground zero at Shot Diablo, and 13,000 feet from ground zero at Shot Shasta.

At Shot Diablo the fallout arrived in the collector between H + 6 minutes and H + 30 minutes.[10] Following the arrival of the fallout, the collector was picked up by a helicopter and flown back to a landing pad near the mobile laboratory trailer containing the equipment for measuring gamma-ray spectra. At the mobile laboratory three samples were prepared from a small portion of the contents of the fallout collector. Sample 1 consisted of about 15 particles which could be picked up with a magnet (see Fig. 1). Sample 2 consisted of five particles, also magnetic. They consisted of melted sand grains coated with a thin layer of a solution of Fe_3O_4 in SiO_2.[11] Thus these samples are not necessarily representative of gross fallout. Sample 3 contained a portion of the non-magnetic residue (also shown in Fig. 1). Samples were put into glass test tubes for measurement.

The sample of fallout material collected after Shot Shasta was returned to NRDL for measurement. A small portion (0.19928 grams total material) of the total fallout material was removed and placed in a test tube for spectral measurement.

GAMMA RAY DETECTION

In the mobile laboratory trailer, gamma-ray spectral characteristics of the samples were observed on a NaI(Tl) crystal scintillation spectrometer and analyzed on a 100-channel pulse-height analyzer. The crystal is cylindrically

shaped, 4 inches high and 4 inches in diameter. The photomultiplier is a 5-inch DuMont type 6364 tube. The 100-channel analyzer is a commercially-available Penco unit. A more detailed description of this equipment is given[12] in WT-1411.

For measurement of gamma-ray spectra, the bottom of each test tube containing the radioactive source was placed on the axis of the aperture of a lead collimator, 16 inches above the bottom of the collimator. The collimator itself was 6 inches long and had a 1/2-inch diameter aperture.

The first run after Shot Diablo was started 92 minutes after the detonation. Sample 1 produced counting rates too high to be handled by the counting equipment, and was not run again until 31 hours after the shot. Sample 3 was not active enough to be analyzed. The gamma rays from Sample 2, containing the smaller number of magnetic pellets, were observed 13 times in the initial 30-hour period after the detonation. The data-taking intervals varied in length from 60 to 300 seconds. The total number of counts recorded in the 100-channel analyzer for a single run varied from 10^5 to 3×10^5.

Three different amplifier gains were used for both Shot Diablo and Shot Shasta data. The high gain setting corresponds to a gamma-ray energy range of 0 - 0.4 Mev. The medium gain corresponds to 0 - 1.8 Mev and the low gain corresponds to 0 - 3.0 Mev.

ANALYSIS

An energy calibration was obtained for each gain setting. Na^{22} and Hg^{203} were used as standards. A typical energy versus channel number curve is shown in Fig. 2.

The pulse-height spectrum from a typical run is shown in Fig. 3. In order to estimate the relative intensity of one gamma ray, say the 1.4 Mev gamma ray, it is necessary to find the area under the total-absorption peak. The pulse-height spectrum from a monoenergetic gamma-ray emitter, taken with the same apparatus, is shown in Fig. 4 for comparison.

The areas for the Shot Diablo data were found using a planimeter. Since the baselines of all but the highest energy peak are above zero, these baselines must be estimated. For most of the runs, the baselines were estimated using the known peak to total ratios as a function of energy. In some cases for Shot Diablo and for the one set of data from Shot Shasta, the spectra were "unfolded" by subtracting off the continuous part of the pulse-height spectrum for each successive gamma ray. This method of analysis is quite tedious and does not increase the accuracy enough to warrant doing it for every run.

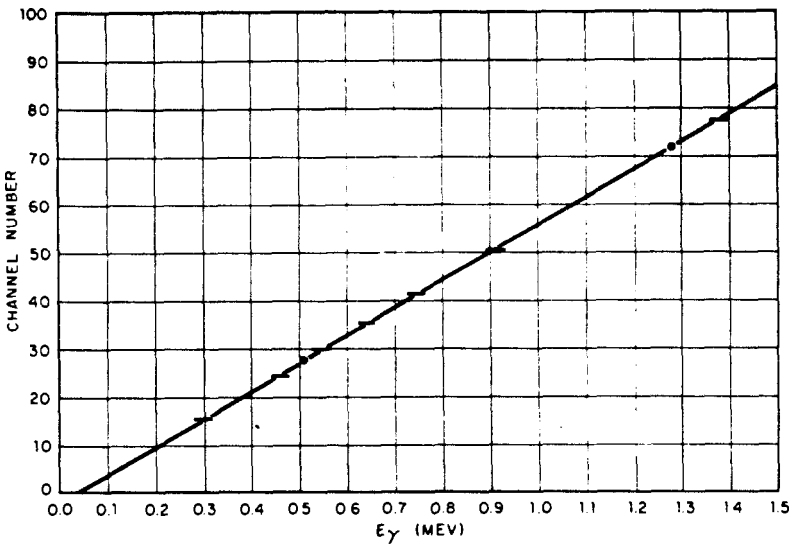

Fig. 2 Gamma-ray energy versus pulse height. The two points are the annihilation quanta and gamma ray from Na^{22}. The horizontal lines are some of the peaks from the pulse-height spectrum of Sample 2 at H + 105 minutes. (See Fig. 3)

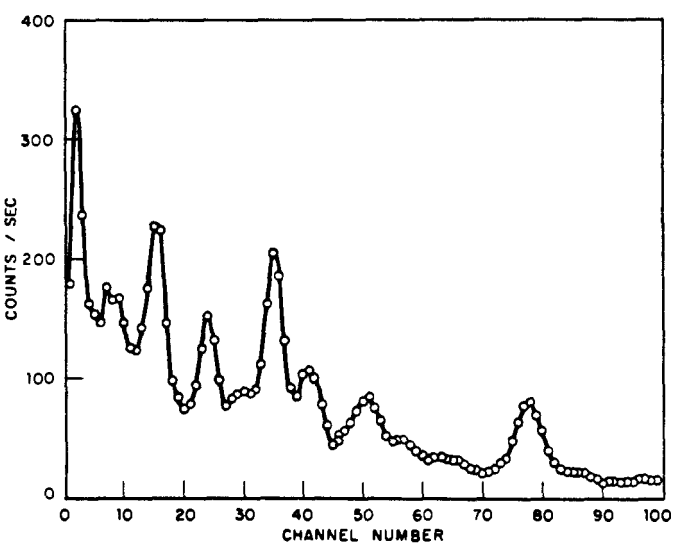

Fig. 3 Pulse-height spectrum of Sample 2 at H + 105 minutes, taken with a 4 x 4 NaI(Tl) crystal and a 1/2-inch lead collimator. See Fig. 2 for an energy calibration. This run was made at the medium gain setting.

To calculate the relative intensity of the gamma rays from the area of the total-absorption peak, it is necessary to know the detector efficiency as a function of gamma-ray energy. The efficiency is a product of the following quantities:

1. Peak to total ratio (from reference 2)

2. Crystal interaction probability; $(1 - e^{-\mu x})$ correction (from reference 2)

3. Collimator edge penetration correction (from reference 13).

The relative efficiency as a function of energy is plotted in Fig. 5.

RESULTS

Table I shows the relative intensity of gamma rays from the samples obtained after Shot Diablo. All gamma rays with measurable relative intensities are shown. The absolute values for the intensities of these gamma rays are of no significance, since they are from an arbitrary sample. Hence the intensities are given in relative numbers only rather than being converted to absolute intensities.

Table II gives the relative intensities of the most prominent gamma rays from the sample obtained after Shot Shasta.

The intensities are divided into groups according to their estimated accuracy. These are denoted by the letters after each intensity. The letter (a) indicates a probable error of 5 to 10 percent, (b) from 10 to 20 percent, and (c) from 20 to 50 percent.

INTERPRETATION

A better knowledge of how the gamma radiation from fallout varies with time and energy may be obtained if the nuclides producing the principal gamma rays can be identified. Then one may use the available decay scheme data to extrapolate in time.

Since no radiochemistry was done on these samples, identification must be made on the basis of gamma-ray energy and half period. Even after the energy and

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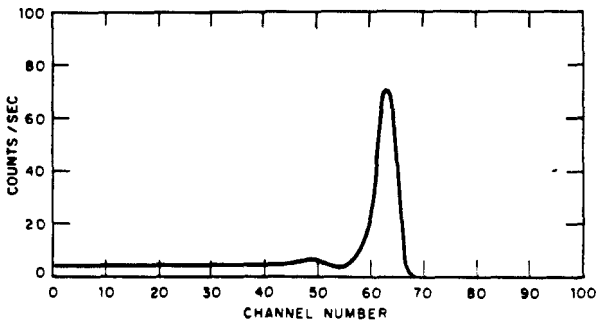

Fig. 4 Pulse-height spectrum of the 1.12-Mev gamma ray of Zn^{65}, taken under the same conditions as in Fig. 3. The annihilation radiation contribution from Zn^{65} has been subtracted.

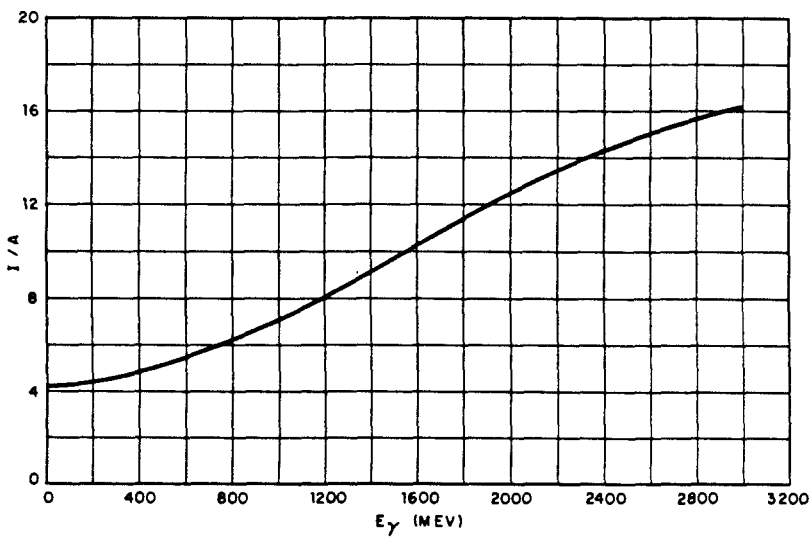

Fig. 5 Relative efficiency for total absorption of gamma rays, I/A, as a function of energy. The numbers on the ordinate are arbitrary, and give the efficiency only relative to other energies, rather than an absolute efficiency.

TABLE I - The relative intensities (number of gamma rays per unit time) of the observed gamma rays from the Diablo data are listed as a function of energy and time. The units are arbitrary, and only give the intensities relative to other numbers in the table. They have been corrected for the variation of the crystal efficiency with energy. The energies are in kev and the times are in minutes after the detonation. The letters following the intensities indicate the accuracy of the intensity determination. A symbol (a) means an accuracy of 5 to 10 percent, (b) means an accuracy of 10 to 20 percent, and (c) means 20 to 50 percent.

The gamma ray energies are accurate to within about 10 kev. In some cases, however, the energy of a given peak changes with time (see Fig. 6). An energy drift down from the quoted energy is indicated by *. An energy drift up from the quoted energy is indicated by ⁄. The symbol ‡ means the peak is not well enough resolved to make a definite energy assignment. The symbol (>0) means that there is evidence of a peak at the indicated energy, but it is too weak for an area measurement.

As explained in the text, three different amplifier gains were used in obtaining the pulse-height spectra. The data taken with low, medium, and high gain runs are designated in the table by l, m, and h, respectively.

	Gain	E (kev) → TAS (min)	16	32	60	73	105	140	170	200	220	240	300
Sample No. 1	m	596.5					72.1c	19.4c			31.1b		61.8b
	m	1846					53.0c	28.0c			14.2b		30.5b
	h	1860	2.61c	16.2b	6.90c	>0	45.0b	28.4b	>0		14.5c		30.3b
	m	1866					55.1c	32.2c			14.7b		31.2b
	m	3953					29.1b	19.2b			7.80b		18.9a
	h	3964	2.51c	8.56b	2.74c	>0	30.8a	17.8a			8.38b		15.8b
Sample No. 2	l	96							58.4c				112 b
	m	105				62.2c	19.1c		30.3c		23.3c		74.6b
	h	117		25.5c		28.6c	13.0c		‡16.2c	‡13.7c		6.91c	37.5c
	l	123.5							39.8c				64.8b
	m	130				39.2c	12.7c	‡27.5c			18.9c		52.2b
	h	137		13.3c		14.7c	9.01c		‡9.71c	‡7.55c			30.3c
	l	168							23.0b				28.9b
	m	175.5				15.9c	21.2c		15.7c		13.9c		26.8b
	h	183		9.41c		3.81c	9.11c		6.17c		6.90c	>0	‡17.2c
	h	191.5		10.4c	>0	2.64c	7.84b	>0	4.97c		5.00c	>0	‡12.8c
	m	568.5			3.78c		12.5c	5.39c	1.10c		4.44c		8.74b
	l	577					14.0b				3.56c		11.86c
	h	590		5.43b	>0		7.27b	2.88b				>0	8.03c

TABLE I (Continued)

	Gain	TAS (min) \ E (kev)	340	450	550	650	740	840	900	1020	1140	1270	1380
Sample No. 1	m	596.5	17.6c	17.7c	59.4b	140 a	145 a		37.0b	28.0b	6.09c		65.1a
	m	1846	5.87c		*15.9b	57.8a	66.9a		--	7.17b	2.23b	>0	2.75c
	h	1860	4.85c										
	m	1866	5.41c		*16.2a	60.2a	67.3a		>0	7.30b	2.23b	1.09c	2.07c
	m	3953	3.99b		* 6.37a	16.3a	22.8a			*1.21c	0.52c	>0	*0.733c
	h	3964	>0										
Sample No. 2	l	96		58.3b	22.9b	108 b	67.9b		72.1b	20.6c	12.2c		111 a
	m	105		46.8a	16.2c	84.6a	47.8b		52.7a	13.1c	18.3c	>0	98.8a
	l	123.5		33.4b	20.1b	90.5b	57.4b		39.4b	25.4c	*9.76c		87.7a
	m	130		28.4a	17.3b	84.6a	43.2b		42.1b	18.1b	8.94c		86.8a
	l	168		12.2b	13.0b	73.0a	51.2a		24.6b	18.0b	8.94a		67.5a
	m	175.5		12.2b	15.4b	67.1a	43.2a	>0	23.5b	13.8b	4.07c		66.5a
	h	183	*2.00c										
	h	191.5	0.94c										
	m	568.5	1.88c	1.76c	8.62b	27.7a	24.7a	>0	5.78b	4.49b	1.01c		*13.5b
	l	577	>0	>0	4.58b	28.3a	21.6a		5.96b	5.99c	*0.80c		*13.5b
	h	590	1.55c										

	Gain	TAS (min) \ E (kev)	1530	1590	1650	1750	1900	2050	2160	2260	2380	2530	2680
Sample No. 1	m	1846		1.90c	0.63c								
	m	1866		1.88c									
	m	3953		3.32a									
Sample No. 2	l	96	10.8c		8.84c	15.9c	19.2c	18.8c	15.3c	7.56c	2.47b	17.2c	3.39c
	m	105	11.9c										
	l	123.5	11.9c		5.83c	10.6c	14.7c	15.0c	13.7c	3.42c	19.4c	15.2c	*3.39c
	m	130.0	>0										
	l	168	4.34c		2.72c	5.70c	8.90c	9.93c	6.42c	1.88c	14.1c	8.62c	1.62c
	m	175.5	>0										
	l	577			*0.66c	*0.79c	*1.22c	*0.64c	*0.70c	0.13c	0.70c	*0.41c	0.16c

TABLE II - The relative intensities of the observed gamma rays from the Shasta data are listed with their energies. These intensities are on a different scale from those in Table I, and are not comparable.

TAS = 3120 minutes

E (kev)	Relative Intensity
30	153 a
55	61 a
100	405 a
135	166 a
225	47 a
285	114 a
520	96 b
662	241 b
745	250 b
960	9.88b
1040	16.4 b
1130	9.55b
1280	5.51c
1380	10.5 c
1590	23.8 c

half period have been determined, within the limit of experimental error, there are often several possible choices. However, the number of possibilities may be decreased by making the following very reasonable assumptions.

1. All the activities observed are U^{235} fission products or radioactivity induced in device materials, such as Np^{239}.

2. The prominent gamma-rays are due to nuclides whose fission yield is at least 1 percent. The tables of Katcoff[14] were used here.

The procedure for identifying a single gamma-ray line in the energy spectrum with its parent nuclide is as follows.

The energy of the gamma ray is carefully determined, using the energies of the radiations from Na^{22} as standards. An energy versus pulse-height curve is shown in Fig. 2. The Na^{22} standard was run several times during the counting, in order to recalibrate the instruments.

Some of the peaks in the pulse-height distribution are due to two or more gamma-rays emitting isotopes having different half lives. If these gamma rays have slightly different energies, a shift is observed as a function of time in the pulse height of the total-absorption peak relative to the peaks of other gamma rays. If all total-absorption peaks in two runs separated by an interval of time are associated with the same set of gamma rays, a plot of the channel position of the total-absorption peaks in one run versus the channel positions of the total-absorption peaks in a later run should all fall on a straight line even though a shift in gain has taken place in the amplifier. If there is a shift in energy of one peak, it is no longer located on this line. Such a shift is seen in Fig. 6. An energy difference of 10 or 20 kev may be detected in this manner. For example one gamma-ray peak in Fig. 6 can be seen shifting from channel 29.5 to channel 28.5 between run number 26 and run number 30. This corresponds to a shift from 550 kev to 535 kev between 596 and 1846 minutes after the shot.

After the energy determination has been made, the relative intensity of the gamma ray is plotted as a function of time. Figure 7 shows one of these graphs. A gamma ray from a single nuclide should appear as a straight line and a half period determination may then be made. If it is not linear (within the limits of experimental error), it means that either the line is a mixture of gamma rays from two or more nuclides, or it is a gamma ray from a nuclide being formed during the time interval of measurement by a radioactive parent.

After the energy and half life determinations have been made, a search is made for a nuclide with the appropriate properties.[15] This is done by picking a time when the gamma ray under consideration is a relatively important part of the spectrum, listing the 10 or 20 most important nuclides at this time according to Bolles and Ballou,[3] and matching properties. If one is found with the correct gamma-ray energy and half life, a check is made on other properties (accompanying

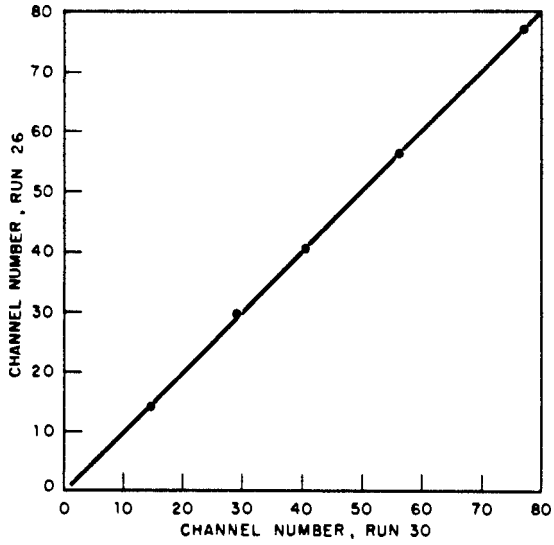

Fig. 6 The peak channel numbers of some of the most prominent gamma rays are shown here for two different runs. The run shown on the abcissa was made at H + 590 minutes, and the run shown on the ordinate was made at H + 1840 minutes; see Fig. 2 for an energy calibration. Note that the peak at about channel 29 shifts to a lower channel number with time, indicating it is a composite of two or more gamma rays.

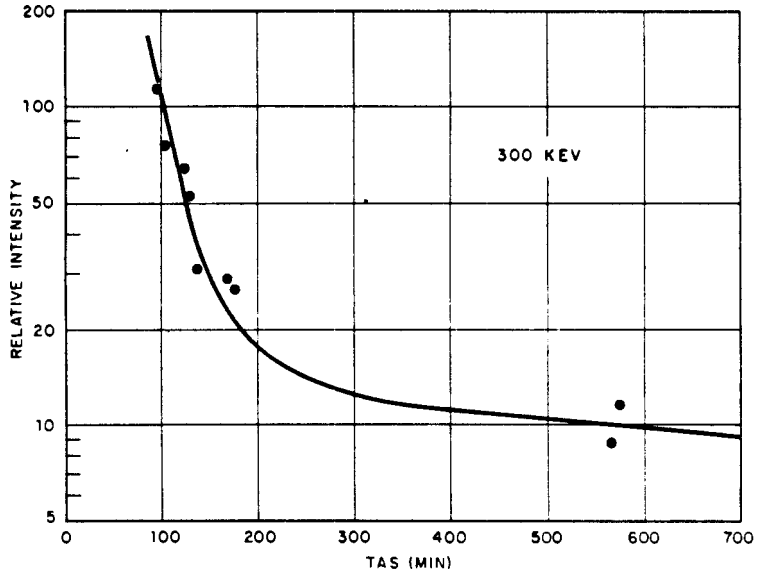

Fig. 7 Relative intensity versus time after shot for the 300-kev gamma-ray line for Sample 2. A short-lived and long-lived component are clearly seen.

TABLE III - This table shows the most probable nuclides from which the observed gamma rays were emitted. The gamma-ray energy given in the table is the one observed in our data, but the half period given is taken from the literature. The numerals following each nuclide denote the degree of confidence the authors have in the assignment. A numeral I indicates that the decay scheme is well understood and agrees well with the data; the numeral II indicates there is an inconsistency between the data and the assignment, but there is not enough accurate data to be positive; and the numeral III indicates that the assigned nuclide is only a "best guess" of the gamma-ray source.

Observed E (kev)	Assigned Nuclide	Reported Half Period
105	Np^{239} (II) (x-ray)	2.3 days
140	Mo^{99} (I)	67 hours
220	Te^{132} (III)	77 hours
300	Ce^{143} (II) Tc^{101} (I)	33 hours 15 minutes
450	Pr^{146} (II)	24 minutes
550	I^{133} (II) Sr^{91} (II)	21 hours 9.7 hours
650	Nb^{97} (I)	72 minutes
740	Zr^{97} (I) Mo^{99} (I)	17 hours 67 hours
900	Y^{92} (I)	3.6 hours
1020	Sr^{91} (II)	9.7 hours
1380	Sr^{92} (II)	2.7 hours
1590	La^{140} (I)	40.2 hours

gamma rays, branching ratios, chemical properties, genetic relationships) and if it agrees, the assignment is made. If the gamma ray is not identified in this way, a further effort is made to find the nuclide by searching for it in Blomeke's list of fission products arranged in order of decreasing half life. In this way gamma rays were identified.

Table III lists the identified gamma rays. The nuclides listed are the negatron-emitting parents, rather than the daughters from which the gamma rays are actually emitted. The relative intensities are extrapolated back to the time of the shot. The Roman numerals indicate the confidence in the assignment by the authors. A numeral I indicates that the decay scheme is well understood and agrees well with the data; the numeral II indicates there is an inconsistency between the data and the assignment, but there is not enough accurate data to be positive; and the numeral III indicates that the assigned nuclide is only a "best guess" of the gamma-ray source.

Approved by:

A. Guthrie

A. GUTHRIE
Head, Nucleonics Division

For the Scientific Director

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