Analysys of Fission Products of Nuclear Explosion by Gamma Ray Spectroscopy

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A very large nuclear explosion of 60 megaton scale [5000 times of 12 kiloton Hiroshima type nuclear weapon] was detonated in Soviet Union on October 29, 1961(1). The fallout particles were blown off high altitude of the earth.

The fission products were gathered by a collector mounted on a jet fighter F-86 F at an altitude of 12 km Gifu Prefecture.

The sample collected from 5 kg of air was measured by a 512 channel gamma ray spectrometer with a NaI (Tl) scintillator of $1\frac{3}{4}'' \phi \times 2''$ size on Decemer 1, 1961.

By the analysis of gamma ray spectra, the energy contribution of the fission of U-238 was estimated approximately 3 times as that of U-235.

I. Introduction

On March 1, 1954 a thermonuclear explosion was detonated on an island in Pacific Ocean. A Japanese fishing boat [Daigo Fukuryu Maru] was drenched by the fission products of uranium. On April 1, the radioactive dose rate was measured by Tatsusaburo SUZUKI, and was obtained the average value at a height of 1 meter on the deck as 0.3 millisievert per hour.

A great number of fission nuclei and induced radioactive elements of U-238 were detected by professor Nobufusa SAITOH of Tokyo University chemically.

The energy contribution of the fission of U-238 of the explosion in 1954 was estimated as 80 percent.

For the purpose of studying the constitution of thermonuclear weapons, it is important to know the contribution of the fission of U-238.

In this study the fission rate of U-238 to U-235 of the detonation on October 29, 1961 was calculated by means of the gamma ray spectroscopy, and its peak efficiencies.

II. Results

The fission products of the nuclear explosion on October 29, 1961 were collected at an altitude 12 km of Gifu Prefecture on November 10, and were measured on

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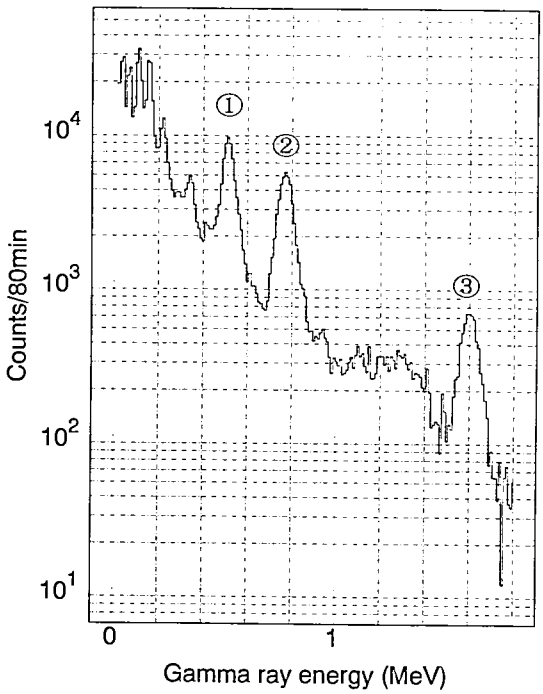

Fig. 1 Gamma ray spectrum of the collected sample.

Table 1. Gamma ray photons of large peaks

Peak (Peak area counts/80min)	Nuclide (Half-life)	Gamma ray energy (MeV)	Branch ratio (%)
Peak ① (42526)	^{103}Ru (39.35d)	0.44380	0.32
		0.49708	90.1
		0.55704	0.82
	^{140}Ba (12.75d)	0.42370	3.12
		0.43755	1.93
		0.53727	24.4
	^{140}La (40.27h)	0.43253	2.99
		0.48703	45.9
	^{147}Nd (11.98d)	0.43990	1.20
		0.48924	0.15
		0.53102	13.1
		0.5896	0.04
		0.59480	0.27
Peak ② (3.68×10^4)	^{95}Zr (64.0d)	0.72418	43.1
		0.75672	54.6
	^{95}Nb (35.0d)	0.76579	99.8
	^{140}La (40.27h)	0.75183	4.41
		0.81580	23.6
		0.86782	5.59
Peak ③ (5400)	^{140}La (40.27h)	1.59649	95.4

December 1, 1961.

The sample collected from 5 kg of air was mounted at a distance of 1.68 cm from the surface of cylindrical NaI (Tl) crystal [$1\frac{3}{4}''\phi\times 2''$] connected to the Nuclear Data 512 channel pulse height analyser.

The counting rate was measured 80 minutes and the gamma ray energy spectrum is shown in Fig. 1. There are 3 large peaks in Fig. 1, peaks ①, ② and ③. The full energy peak area[2] of each peak and the corresponding nuclei are shown in Table 1.

The radioactivity of ^{140}La in Table 1 is determined from the area of peak ③ and its efficiency, where the full energy peak efficiency η is defined as the following equation

$$\eta=\frac{\text{number of pulses recorded in peak}}{\text{number of photons emitted from source}}. \tag{1}$$

The full energy peak efficiencies of standard gamma rays were measured by Tatsuo URAI[3] and is shown in Table 2.

Assuming the relation between η and gamma ray energy $h\nu$ is represented as

$$\log\ \eta=a\ \log\ (h\nu)+b, \tag{2}$$

the η of peak ③ for 1.59649 MeV gamma radiation from ^{140}La may be obtained by extrapolation of the values in Table 2 as

$$\eta\,③=0.0055. \tag{3}$$

As the branch ratio of 1.59649 MeV gamma rays from ^{140}La is 0.954, the radioactivity of ^{140}La is determined from Table 1 and Eq. (3) as

$$A\,(^{140}\text{La})=5400/4800\text{sec}/0.0055/0.954=214.4\ \text{Bq}. \tag{4}$$

Since the ^{140}La nuclide is the daughter element of ^{140}Ba, the ratio of the radioactivity of ^{140}La to that of ^{140}Ba after 32 days (Oct. 29-Dec. 1) of fission is determined from half-lives as

Table 2. Full energy peak efficiency η for standard gamma radiation[3]

Nuclide	Gamma ray energy (MeV)	η
^{22}Na	1.27454	0.00718
^{54}Mn	0.83483	0.0118
^{137}Cs	0.66165	0.0165
^{133}Ba	0.35600 0.30285	0.0359 0.0775

$$A(^{140}\mathrm{La})/A(^{140}\mathrm{Ba})=1.1515, \tag{5}$$

then the radioactivity of ^{140}Ba is obtained as

$$A(^{140}\mathrm{Ba})=186.2\ \mathrm{Bq}. \tag{6}$$

Ⅲ. Number of Photons of Peak ①

The counting rate of peak ① in Fig. 1 is obtained 42526 c/80 min as is shown in Table 1. From Fig. 1 the annihilation radiation ($h\nu=0.511$ MeV) of positrons from 1.6 MeV gamma radiation (peak ③) is included.

As the contribution of 0.511 MeV radiation is about 1% of the area of peak ③[2], the net counting rate from ^{103}Ru, ^{140}Ba, ^{140}La and ^{147}Nd is 4.247×10^4c/80 min.

The η of peak ① is obtained from Table 2 by interpolation as

$$\eta①=0.027, \tag{7}$$

then the total number of photons from ^{103}Ru, ^{140}Ba, ^{140}La and ^{147}Nd is obtained as

$$N_T = 1.573\times10^6/80\ \mathrm{min} = 327.7/\mathrm{sec}. \tag{8}$$

The number of gamma radiation in the energy region peak ① (0.4 MeV-0.6 MeV) from ^{140}Ba and ^{140}La is obtained from Eqs. (4) , (6) and Table 1 as 54.84/sec and 104.82/sec respectively.

The sum of the number of photons emitted from ^{103}Ru and ^{147}Nd in the energy region peak ① in unit time is obtained from Eq. (8) as

$$N_1 = 168.04/\mathrm{sec}. \tag{9}$$

The experimental ratio of N_1 to N_T is

$$R_E = N_1/N_T = 0.513. \tag{10}$$

Ⅳ. Calculation

The relative fission yield α_γ of U-235 and U-238 by fast neutrons is shown in Table 3[4].

The number of photons from each parent nucleus may be represented as

$$N_i = \lambda_i n_i\ \alpha_{bi} = \lambda_i n_{i0}\ e^{-\lambda i t}\ \alpha_{bi}, \tag{11}$$

where

λ : decay constant

α_b : gamma photon branch ratio

n_i : number of atoms of measured time

n_{i0} : number of atoms of fission time.

As the number of atoms of fission time is proportional to the yield ratio α_γ, n_{i0} in

Table 3. Relative fission yield α_y by fast neutron[4]

Mass unmber A	α_{y1} for U-235	α_{y2} for U-238
A=103	3.0	6.0
A=140	6.0	6.0
A=147	2.0	2.4

Eq. (11) is represented as

$$n_{i0} = \alpha_y n_0, \tag{12}$$

where n_0 is the proportionality constant. Assuming the number of the fission of U-238 atoms to that of U-235 atoms is x, the fission yield may be shown as

$$\alpha_y = \alpha_{y1} + x\alpha_{y2}, \tag{13}$$

and from Eq. (11) the radioactivity is written as

$$A = \lambda_i n_i = \lambda_i \alpha_y n_0 e^{-\lambda i t}. \tag{14}$$

Since the number of photons of the energy of peak ① emitted from ^{140}Ba and ^{140}La is calculated from Table 1, Table 3, Eqs. (5) and (14) as

$$\begin{aligned} N_{C2} &= A(^{140}\text{Ba})\,\alpha_b(^{140}\text{Ba}) + A(^{140}\text{La})\,\alpha_b(^{140}\text{La}) \\ &= A(^{140}\text{Ba})\left[\alpha_b(^{140}\text{Ba}) + \frac{A(^{140}\text{La})}{A(^{140}\text{Ba})}\,\alpha_b(^{140}\text{La})\right] \\ &= A(^{140}\text{Ba})\,[0.2945 + 1.15153 \times 0.4889] \\ &= 0.8575A(^{140}\text{Ba}) \\ &= 0.8575(6+6x)\,\frac{\ln 2 \cdot n_0}{12.75\text{d}}\,e^{-(\ln 2/12.75)\times 32} \\ &= 0.04911(1+x)n_0/\text{d}. \end{aligned} \tag{15}$$

On the same way, the number of photons from ^{103}Ru and ^{147}Nd of energy region peak ① is

$$\begin{aligned} N_{C1} &= \ln 2 \cdot \left[\frac{1}{39.35\text{d}}\,(3+6x)e^{-(\ln 2/39.35)\times 32} \times 0.9124\right. \\ &\quad \left. + \frac{1}{11.98\text{d}}\,(2+2.4x)e^{-(\ln 2/11.98)\times 32} \times 0.1476\right] \\ &= (0.030122 + 0.0581x)n_0/\text{d}. \end{aligned} \tag{16}$$

From Eqs. (15) and (16), the total calculated number of photons from ^{103}Ru, ^{140}Ba, ^{140}La and ^{147}Nd in the energy of peak ① is

$$N_{TC} = (0.07923 + 0.10721x)n_0/\text{d}, \tag{17}$$

and the calculated ratio of N_{C1} to N_{TC} may be represented as

$$\frac{N_{CI}}{N_{TC}} = \frac{0.03012+0.0581x}{0.07923+0.10721x}. \tag{18}$$

V. Fission Rate of U-238 to U-235

As the calculated ratio in Eq. (18) should be equal to that of experimental result in Eq. (10) , the fission rate in Eq. (13) may be obtained from the following equation

$$\frac{0.03012+0.0581x}{0.07923+0.10721x} = 0.513 \tag{19}$$

or

$$x=3. \tag{20}$$

A large number of fast neutrons are produced by the fission of uranium or by the fusions of ^{2}D or ^{3}T in the thermonuclear explosive reaction. Many of them will initiate the fissions of uranium, (n, γ) reactions or $(n, 2n)$ reactions directly or after elastic collisions in a very short time ($\sim 10^{-5}$ sec) , and some part of them perform the nuclear explosive chain reactions.

For 3F (Fission → Fusion → Fission) type nuclear explosive device of megaton scale, several tons of uranium will be prepared. Highly enriched (more than 90 percent U-235 atoms) uranium blocks are located in the center of the device as an igniter and about ten times of the weight of natural uranium is arranged outside of the device as a tamper.

Nevertheless the fission cross sections for both U-235 and U-238 are about the same for fast neutrons[6], some U-235 atoms located in the center of the device will be able to initiate fission by neutrons scattered inelastically in an explosive time and fission capability of U-235 is larger than that of U-238, and in this case it may be considered that the number of the fission of U-238 will be only several times than that of U-235 though the weight of U-238 is about ten times of U-235.

References

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