Fission Product Gamma Spectra

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by

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ABSTRACT

The fission product gamma spectra of ^{233}U, ^{235}U, and ^{239}Pu have been measured at 12 cooling times following 20 000-s irradiations in the thermal column of the Omega West Reactor. The mean cooling times ranged from 29 s to 146 500 s. The total gamma energies were obtained by integrating over the energy spectra, and both the spectra and the total energies are compared with calculations using the CINDER-10 code and ENDF/B-IV data base.

The measured and calculated gamma spectra are compared in a series of figures. The measured total gamma energies are ∼14% larger than the calculated energies during the earliest counting period (4 s to 54 s cooling time). For ^{235}U, the measured and calculated total gamma energies are nearly the same after 1200 s cooling time, and the measurements are 2% to 6% lower at longer cooling times. For ^{239}Pu, the measured and calculated total gamma energies are nearly the same at cooling times longer than 4 000 s, and for ^{233}U this condition prevails at cooling times longer than 10 000 s.

I. INTRODUCTION

The fission product gamma spectra of ^{233}U, ^{235}U, and ^{239}Pu have been measured at 12 cooling times following 20 000-s irradiations in the thermal column of the Omega West Reactor. The radiation period was long enough to build up a near-equilibrium concentration of the shorter half-life fission products. The constant neutron flux was abruptly cut off at the end of each irradiation.
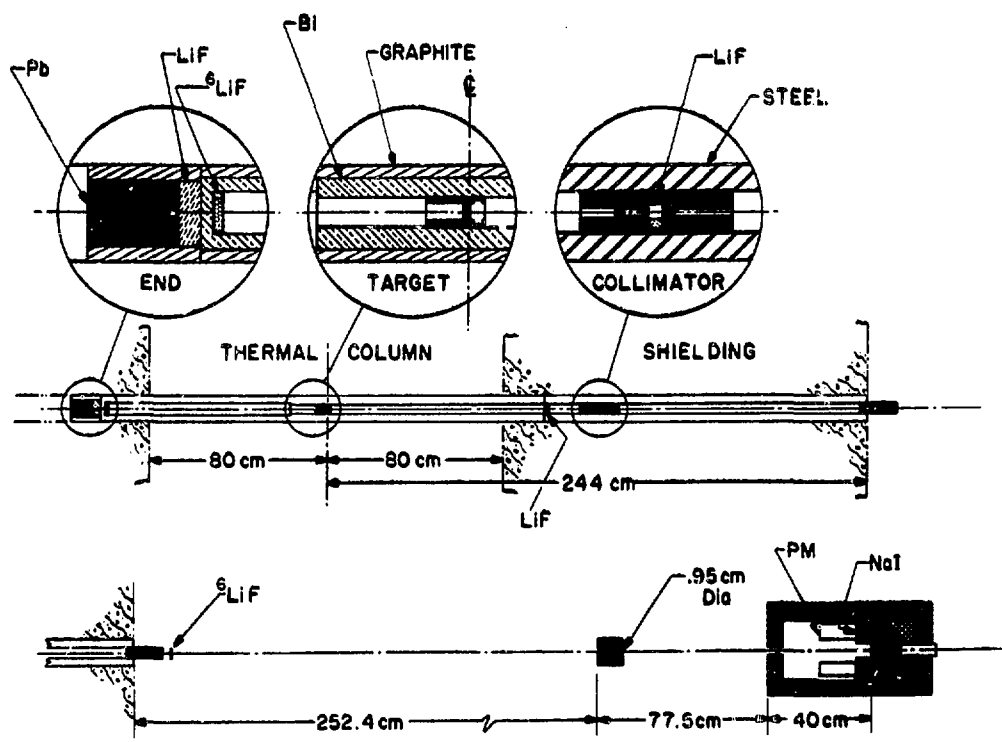
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Fig. 1. Plan view of the gamma spectrometer. The continuation of the beam path outside the reactor shielding is shown below. The graphite holder is shown in the middle circle.

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The present work was initiated in connection with cryogenic calorimetric measurements of the fission product decay heat.[1,2] The gamma and beta spectra from ^{235}U and ^{239}Pu fission products following 1- to 100-s irradiations have been measured by Dickens et al[3,4] at cooling times between 2 and 14 000 s.

II. THE REACTOR AND SPECTROMETER

The Omega West Reactor is a light-water, thermal-neutron reactor using a central core of Materials Testing Reactor type fuel elements containing

enriched uranium. A 76-mm-thick lead shield adjacent to the core and a 127-mm-thick bismuth shield inside the thermal column prevent photons originating in the core from reaching the sample in the thermal column.

A bismuth-walled evacuated channel, in which the sample is placed, extends through the thermal column, as shown in Fig. 1. The bismuth attenuates the equilibrium thermal-column gamma flux at the sample position, and collimation prevents gammas originating in the channel walls from reaching the detector. The sample is located 6.14 m from the detector, and the photon beam is collimated to approximately 1-cm diameter at the detector. The detector was a 6.35-cm-diam by 15-cm-long NaI(Tl) crystal located inside a 25-cm-diam by 30-cm-long NaI(Tl) annulus, used in anticoincidence (total energy mode). The entire detector was enclosed in a 7.6-cm-thick lead and iron shield, as shown in Fig. 1.

During sample irradiations, the reactor power was held constant at 8 MW, corresponding to a thermal neutron flux of 6×10^{11} n/cm^2s at the sample. The thermal flux approximates a Maxwellian distribution corresponding to a temperature of 350 K, for which the most probable neutron velocity is 2 400 m/s.

III. THE SAMPLES

The weight and isotopic composition of the fissionable samples is given in Table I. The ^{233}U and ^{235}U samples were 6.35-mm-diam disks, and the ^{239}Pu

TABLE I

WEIGHT AND COMPOSITION (IN WEIGHT PER CENT) OF THE SAMPLES

Sample	Weight mg	Chemical Composition	Isotope	Isotopic Composition
^{233}U	1.73	U = 100%	U-233	97.46 w/o
			U-234	1.15
			U-235	0.29
			U-236	0.04
			U-238	1.06
^{235}U	2.14	U = 100%	U-234	0.87 w/o
			U-235	93.19
			U-236	0.48
			U-238	5.46
^{239}Pu	8.6	Pu = 99.37 w/o	Pu-238	0.04 w/o
			Pu-239	93.59
			Pu-240	6.02
		Other elements = 0.63	Pu-241	0.32
			Pu-242	0.03

3

sample was a 4.76-mm-diam disk, all three cut from 0.025-mm-thick foils. They were sealed between 0.125-mm-thick aluminum disks cf larger diameter. The aluminum disks holding the uranium foils were electron-beam welded around the perimeter, while the plutonium disk was sealed between aluminum disks with Armstrong epoxy.[*] The samples were carried upright in graphite holders when inserted into the thermal column. No leakage of gaseous fission products was detected by radiation monitors adjacent to the exhaust line from the pump that evacuated the thermal column channel.

Dummy samples consisting of aluminum disks and epoxy where appropriate were also irradiated and counted. The dummy sample spectra were later subtracted from the fissionable isotope spectra.

IV. STRIPPING THE GAMMA SPECTRA

The pulse-height distribution from the detector during a counting period was accumulated in a pulse-height analyzer and read onto magnetic tape for processing by computer. The computer stripping code takes account of the following unwanted features in the response of the NaI(Tl) crystal to a gamma line: (1) bremsstrahlung escape pulses on the low-energy side of the full-energy peak; (2) the Compton edge, a small peak below the full-energy peak; (3) the flat distribution of Compton-scattered photons below the Compton edge; (4) the first and second annihilation radiation escape peaks; and (5) a back-scattering peak, at ∿ 200 keV. The stripping starts at the high-energy end of pulse-height distribution, treating the top bin as a portion of a full-energy peak, and reduces the counts in the lower energy bins by an amount determined by the response function. The process is repeated successively for the lower energy bins.

V. CALCULATING GAMMA POWER FROM THE MEASUREMENTS

Before irradiating ach fissionable target, a 44.2-mg sample of polyethylene $(CH_2)_n$ was irradiated, and the hydrogen capture gamma ray at 2.223 MeV $(=E_H)$ was counted by the spectrometer for a convenient period. The fissionable target was then irradiated in the same neutron flux for 20 000 s, after

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[*]Armstrong Products Co., Argonne Road, Warsaw, IN. Type 1-A resin with activator A was used.

which the reactor was abruptly shut down. This procedure enabled us to calibrate the spectrometer in terms of unit reaction cross section for a constant neutron flux, since the (n,γ) cross section for ^1H is accurately known. The gamma power at cooling time t, $\Gamma(E,t)$ per unit fission rate per unit energy interval about E, is given by

$$\Gamma(E,t) = \frac{G(E,t)E\ \epsilon(E_H)\ N_H \sigma_H(n,\gamma)}{G(E_H)\ \epsilon(E)\ N_U \sigma_U(fiss)} \quad ,$$

where $G(E_H)$ is the gamma counting rate recorded by the spectrometer for the hydrogen capture line, and $G(E,t)$ is the gamma counting rate recorded in the 50-keV energy interval about E. The function $\epsilon(E)$ is the relative efficiency of the spectrometer at energy E. N_H and N_U are the number of hydrogen and uranium (or plutonium) atoms.

Except for ^{233}U, the $\sigma(fiss)$ used in Eq. 1 are not 1/v cross sections. We have therefore integrated the $\sigma(fiss)$ and the neutron absorption cross sections $\sigma(abs)$ over a Maxwellian thermal neutron distribution having a most probable velocity of 2 400 m/s. The $\sigma(abs)$ were used to calculate a correction for self-absorption and for neutron-flux depression at the location of the samples. For those cross sections which are 1/v, the mean velocity of the Maxwellian distribution is $2/\sqrt{\pi}$ times 2 400, which equals 2 708 m/s. The cross sections used are given in Table II, and are based on curves given in BNL-325.[5]

TABLE II

NEUTRON ABSORPTION CROSS SECTIONS AND QUANTITIES OBTAINED BY

SUMMING OVER THE SPECTRA FROM 50 keV TO 7 MeV

Isotope	σ_{abs} barns	Multiplicity γ's/neutron	Spectral Sum barns x MeV	Energy Released MeV/neutron	Average Photon Energy MeV/photon
^{233}U	470	14.6 $\pm$ 0.3	6920 $\pm$ 248	14.7 $\pm$ 0.5	1.01 $\pm$ 0.06
^{235}U	533	13.6 $\pm$ 0.3	6918 $\pm$ 238	13.0 $\pm$ 0.4	0.95 $\pm$ 0.05
^{239}Pu	887	11.2 $\pm$ 0.3	9200 $\pm$ 243	10.4 $\pm$ 0.3	0.92 $\pm$ 0.05
^1H	0.270 [a]	1.0			2.22

[a] The BNL-325 value for 2200 m/s neutron velocity is 0.332 barns.

The results are given as tables of gamma power in MeV per second, per energy interval, at cooling time t, divided by the number of fissions per second during the irradiation. The units MeV per second divided by fissions per second are conventionally called MeV/fission, even though the denominator refers to the fission rate during neutron irradiation and the numerator refers to a rate after the reactor was shut down. Each value is given with an uncertainty, which includes a 1^σ statistical uncertainty in quadrature with a $\pm10\%$ uncertainty assigned to the response function used in stripping the spectra. The spectra have also been summed over the 50-keV energy bins from zero to 6 MeV to obtain the total gamma power at cooling time t.

VI. GAMMA SPECTRA CALCULATED FROM FISSION PRODUCT NUCLIDES

In the next section we include comparisons of the measurements with calculated spectra and with total gamma energy release rates. Calculated spectra are obtained using the CINDER-10 code to generate activities and total gamma decay energies. The activities are folded into multigroup energy spectra using an auxiliary routine which is a modification of FPSPEC.[6] CINDER-10 is not documented but is an extensive modification of CINDER-7.[7] The spectra, fission yields and all decay parameters used in CINDER-10 are processed from Version IV of the fission-product Evaluated Nuclear Data Files (ENDF/B-IV),[8] except for data corrections noted in Ref. (9). There are 824 nuclides in the ENDF/B-IV fission product file, 711 being unstable. Gamma energies and intensities are included for 180 of the unstable nuclides; as will be shown in the next section, this subset contributes most of the total calculated gamma energy for the cooling times covered by the measurements. All unstable nuclides contain a total gamma energy; these and all other ENDF/B-IV fission product parameters (branching fractions, decay constants, fission yields, cross sections, etc.), are incorporated into the CINDER-10 library. The calculations are then made for the specific irradiation history of the fission samples and the spectra subsequent to irradiation are calculated at the mid-point of each counting period. The calculations do not have any significant approximation but are dependent on the completeness and quality of the nuclide data. The comparisons in the next section indicate the quality and completeness of the calculated spectra; this section itemizes the more significant aspects considered in the calculations.

For each of the 180 nuclides having spectra data, the gamma energies are first broadened to match the resolution and energy dependence of the spectrometer using a Gaussian distribution about each gamma energy, E_0, and having an area equal to the intensity per decay, I:

$$G = \frac{I}{\sigma\sqrt{2\pi}} \exp \left[-\left[\frac{(E-E_0)^2}{2\sigma^2} \right] \right] \quad .$$

(In this experiment, $\sigma = -2 + 0.47 \sqrt{E}$, where E is in keV.) The subsequent distributions are then grouped into 150 energy bins on a uniform 50 keV grid between 0 and 7.5 MeV. In this report we have grouped the energies, not the photon multiplicities, per bin. Plots of the resulting spectra then visually display the energy release over the energy axis.

We have assumed that the spectral shape is determined by the 180 nuclides having spectral data in ENDF/B-IV and have normalized the aggregate gamma spectra to provide the total value calculated by CINDER-10.

Measured spectra are based on a finite counting time but calculations apply to specific decay times. For energy-integrated comparisons, we have examined the difference between using a time average of the calculated values over the counting period and the energy release rate at the midpoint of the period. The mid-point value is ∼1% smaller than the time-averaged value. This evaluation was made before the final measurements and did not include the subsequent 4-54 s counting interval. For this period we have calculated the mid-point release rate to be nearly 5% smaller than the time-averaged value. These differences are not removed in the tabular comparisons of the integrated spectra.

VII. RESULTS

The schedule of counting periods following reactor shutdown is given in Table III. The successive counting periods are identified on the figures and in the tables of results by the midpoint values of the periods.

The total gamma energies at the various cooling times are given in Table IV. The last column under each isotope in Table IV gives the per cent of the total calculated energy which is contributed by the 180 nuclides. The results in Table IV apply only to a 20 000-s irradiation at constant power.

TABLE III

SCHEDULE OF COUNTING PERIODS AFTER REACTOR SHUTDOWN

Period Number	Starting Time, s	Duration s	Midpoint: Aver. Cooling Time, s
1	4	50	29
2	68	120	128
3	194	180	284
4	388	300	538
5	918	600	1218
6	2170	720	2530
7	3480	900	3930
8	4560	900	5010
9	22860	1800	23760
10	57970	2700	59320
11	95040	3600	96840
12	144700	3600	146500

TABLE IV

INTEGRATED γ-SPECTRUM: COMPARISON OF LASL MEASUREMENTS AND CINDER-10 PLUS ENDF/B-IV

	^{235}U			^{233}U			^{239}Pu		
COOLING TIME(s)	MeV/FISS EXP	RATIO EXP/CAL	% ENERGY FROM 180 NUCLIDES	MeV/FISS EXP	RATIO EXP/CAL	% ENERGY FROM 180 NUCLIDES	MeV/FISS EXP	RATIO EXP/CAL	% ENERGY FROM 180 NUCLIDES
29	3.762 ± 0.198	1.140	80.4	3.333 ± 0.155	1.131	85.2	3.100 ± 0.168	1.136	74.6
128	2.502 ± 0.108	1.080	91.5	2.430 ± 0.094	1.111	92.9	2.136 ± 0.086	1.084	87.1
284	1.975 ± 0.089	1.057	94.7	1.982 ± 0.083	1.103	95.1	1.747 ± 0.074	1.085	92.2
538	1.653 ± 0.071	1.057	96.2	1.647 ± 0.068	1.087	96.3	1.502 ± 0.061	1.102	94.7
1218	1.195 ± 0.027	0.995	97.6	1.215 ± 0.025	1.052	97.3	1.104 ± 0.024	1.058	96.8
2530	0.873 ± 0.029	0.979	98.9	0.839 ± 0.028	1.022	98.8	0.725 ± 0.025	1.029	98.6
3930	0.626 ± 0.029	0.975	99.5	0.643 ± 0.028	1.024	99.4	0.533 ± 0.024	1.007	99.2
5010	0.531 ± 0.034	0.981	99.7	0.551 ± 0.035	1.036	99.6	0.444 ± 0.028	1.012	99.4
23760	0.108 ± 0.004	0.937	99.8	0.121 ± 0.004	0.983	99.7	0.099 ± 0.003	0.998	99.5
59320	0.042 ± 0.002	0.982	99.9	0.044 ± 0.002	1.024	99.8	0.041 ± 0.002	1.004	99.4
96840	0.024 ± 0.002	0.989	99.8	0.024 ± 0.002	0.990	99.8	0.024 ± 0.002	0.995	99.3
146500	0.014 ± 0.002	0.961	99.8	0.015 ± 0.002	1.013	99.8	0.015 ± 0.002	1.004	99.2

ENDF/B-IV files contain internal electron conversion coefficients for 38 important nuclides. The spectral comparisons do not include these conversion energies and the tabular comparisons of the energy-integrated spectra are given without the internal conversion energies. However, only the conversion energies of the 38 nuclides having internal conversion coefficients in ENDF/B-IV were excluded.

Bendt and Jurney[10] have also made gamma-power measurements (after 20 000 s) while the irradiation of the fissionable isotopes continued. Peelle and Maienschein[8] have measured the prompt photon energy in coincidence with fission fragments, for ^{235}U. Combining these measurements results in a gamma power from fission products at zero time equal to 6.9 ± 0.7 MeV/fission (for
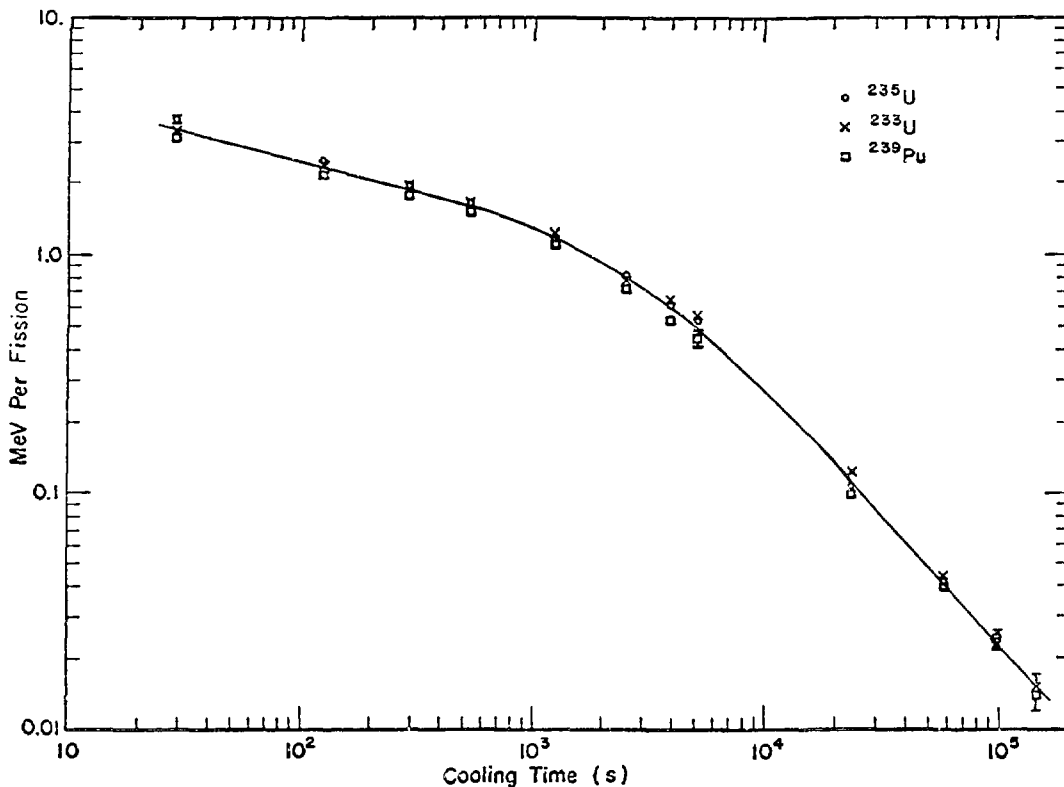
Fig. 2. The experimental gamma power of the fissionable isotopes versus cooling time. Representative error bars are shown on a few points.

^{235}U). This quantity could be obtained for the other isotopes if the prompt photon energy released at the time of fission were measured.

Two figures show the results given in Table IV. Figure 2 shows the experimental total gamma power of the fissionable isotopes plotted versus cooling time. In Fig. 3, the difference between the measurements and calculations, as a percent of the measured values, is plotted against cooling time. The trend shown in Fig. 3, from $\sim$ 12% at 29 s, to values around zero percent and lower, at times longer than 1000 s, is not understood.

Figures 4 through 39 (at end of report) show the measured spectrum in MeV/fission/50 keV bin, and the calculated spectrum as a continuous line, for each isotope and cooling time.

ACKNOWLEDGMENTS

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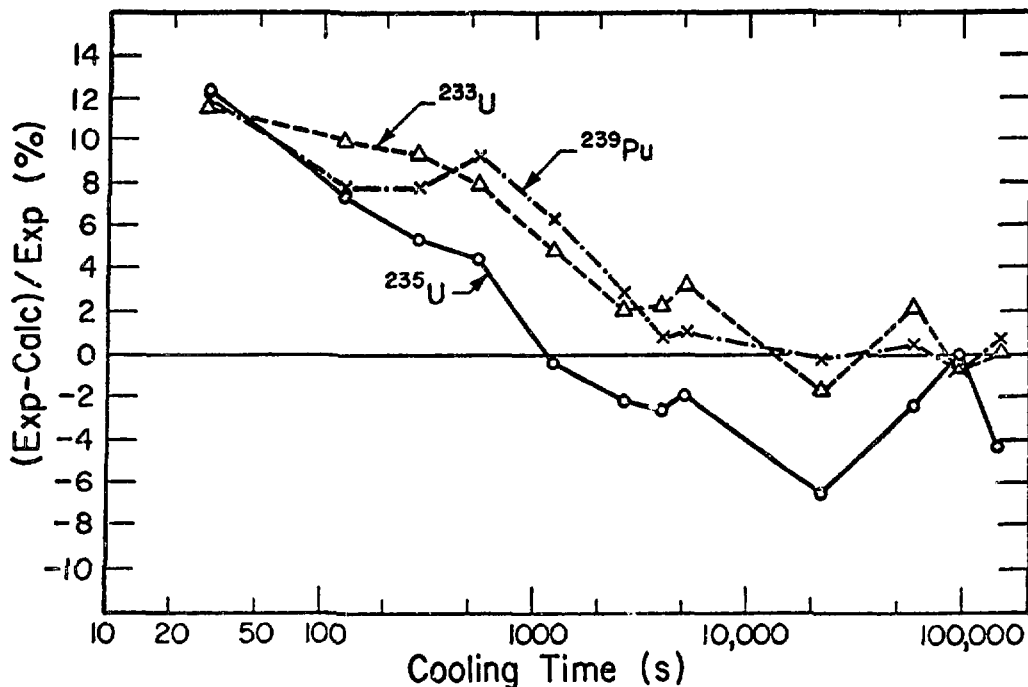
Fig. 3. Difference between the experimental and calculated gamma power, as a percent of the experimental values, versus cooling time.

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10

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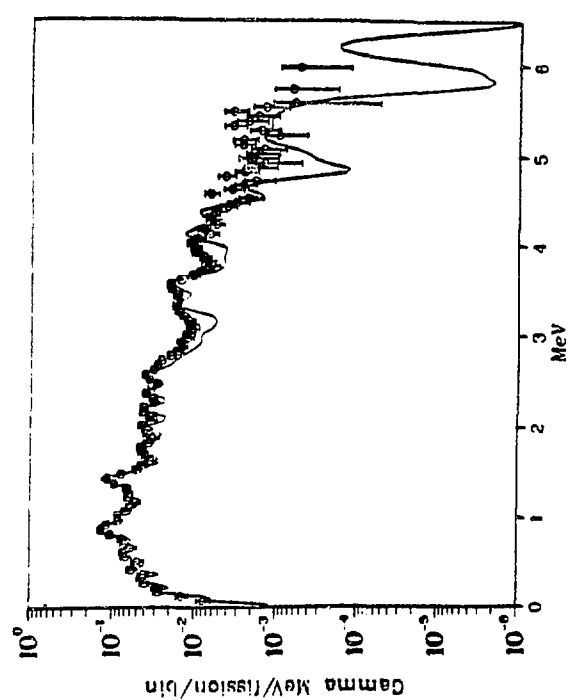
Fig. 4. ^{233}U, after 29 s cooling.

Fig. 5. ^{233}U, after 128 s cooling.

Fig. 6. ^{233}U, after 284 s cooling.
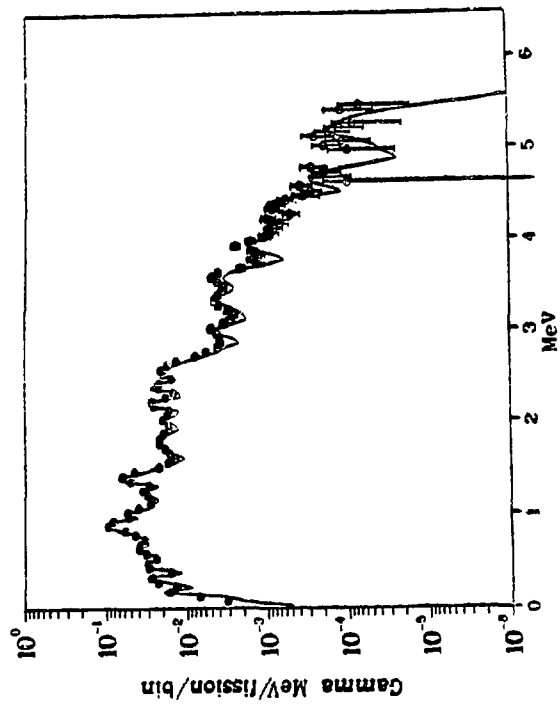
Fig. 7. ^{233}U, after 538 s cooling.

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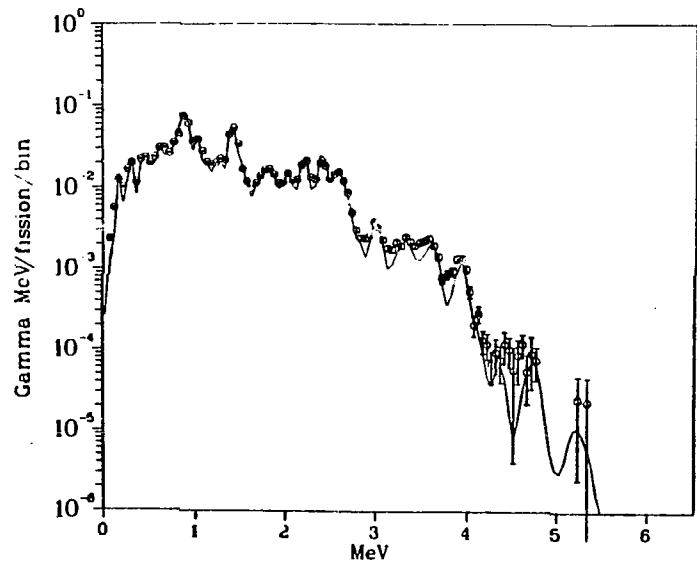
Fig. 8. ^{233}U, after 1218 s cooling.
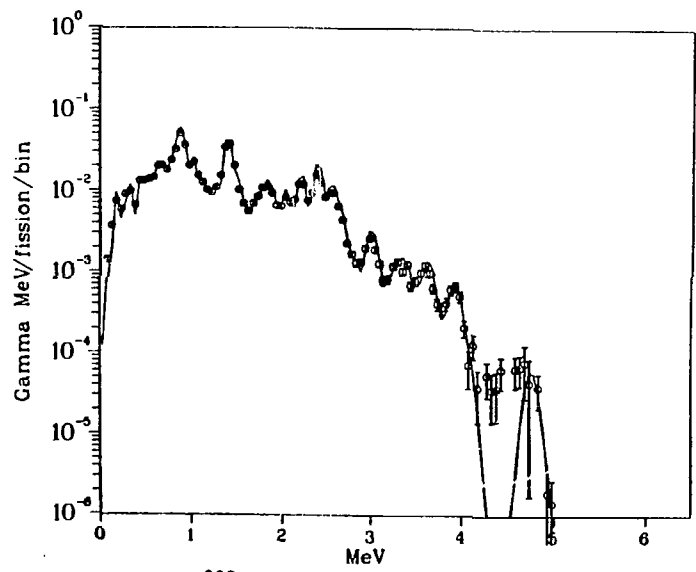
Fig. 9. ^{233}U, after 2530 s cooling.
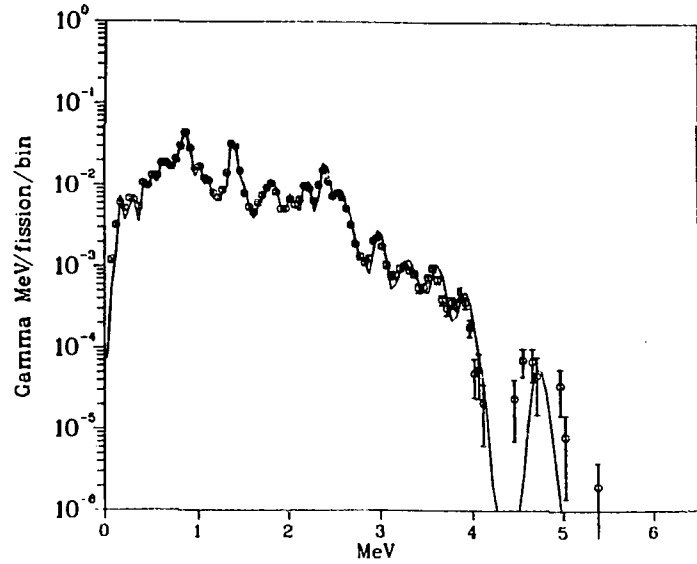
Fig. 10. ^{233}U, after 3930 s cooling.
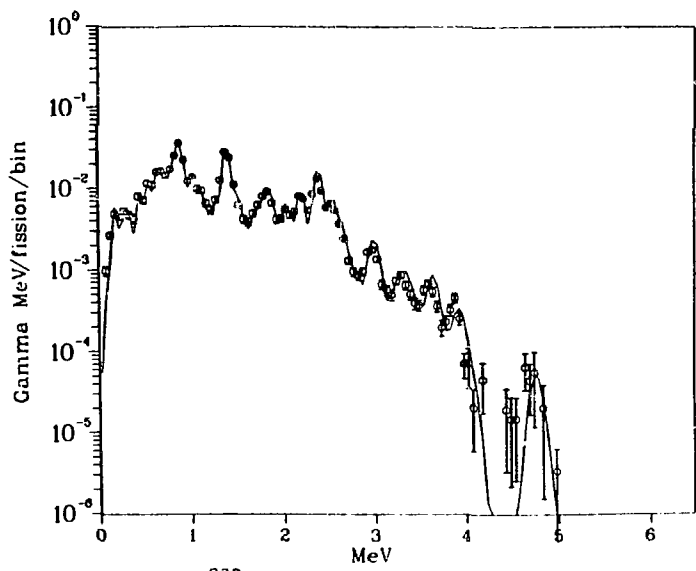

Fig. 11. ^{233}U, after 5010 s cooling.
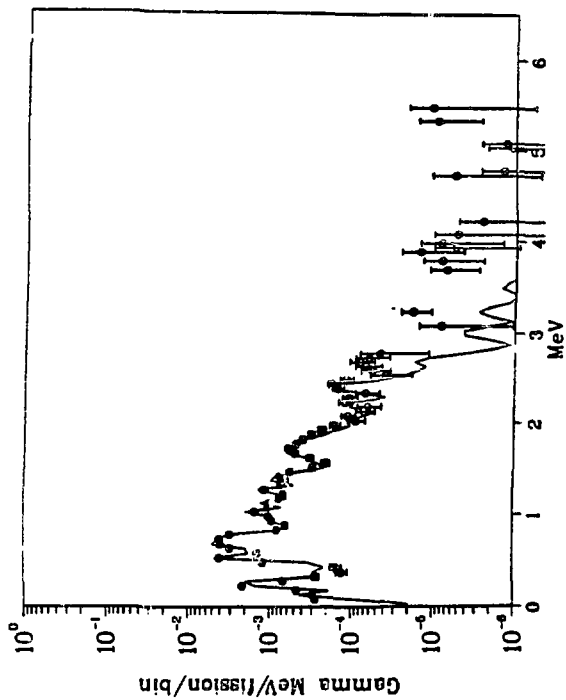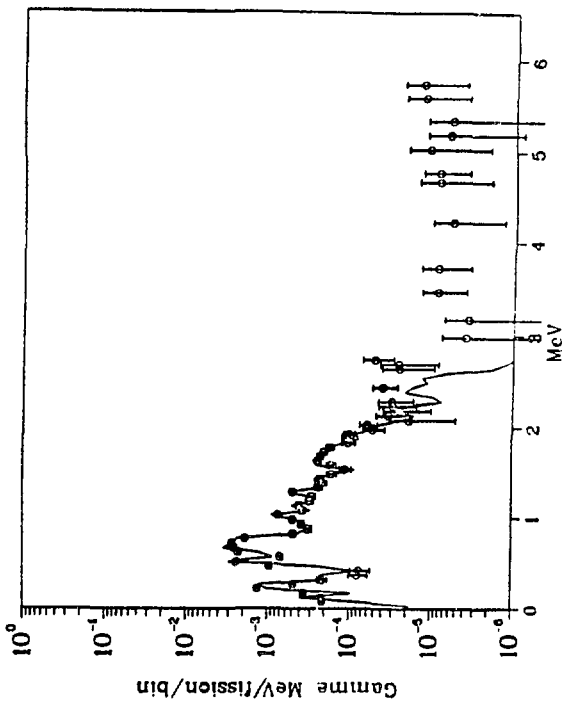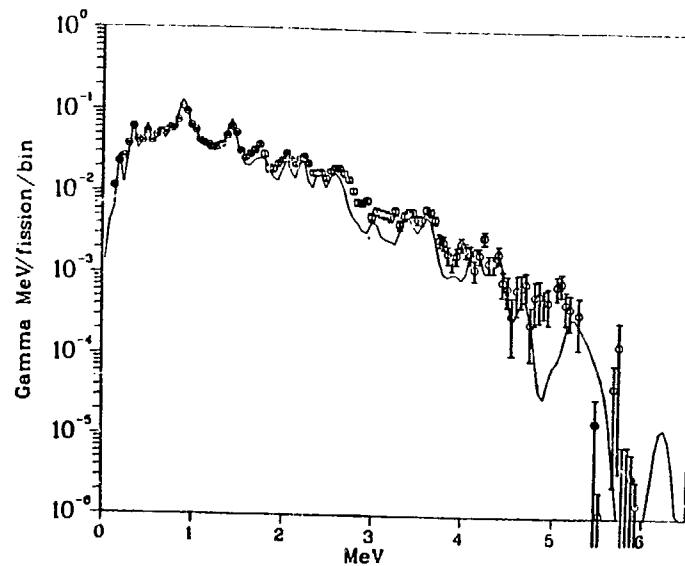
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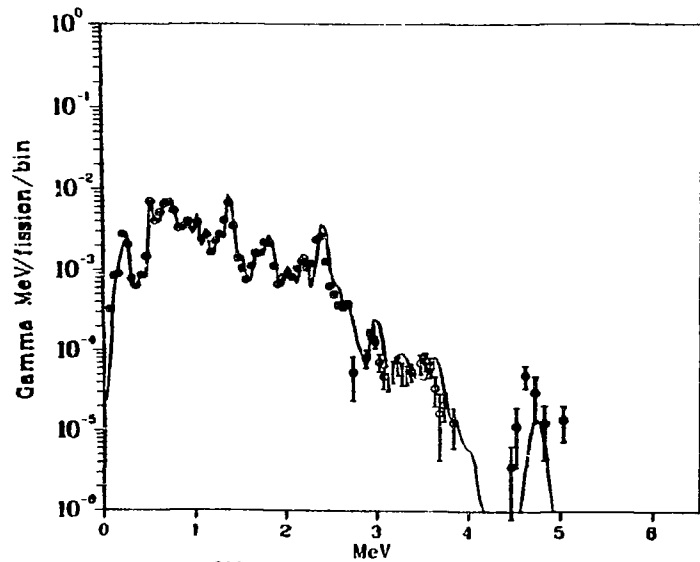
Fig. 12. ^{233}U, after 23 760 s cooling.
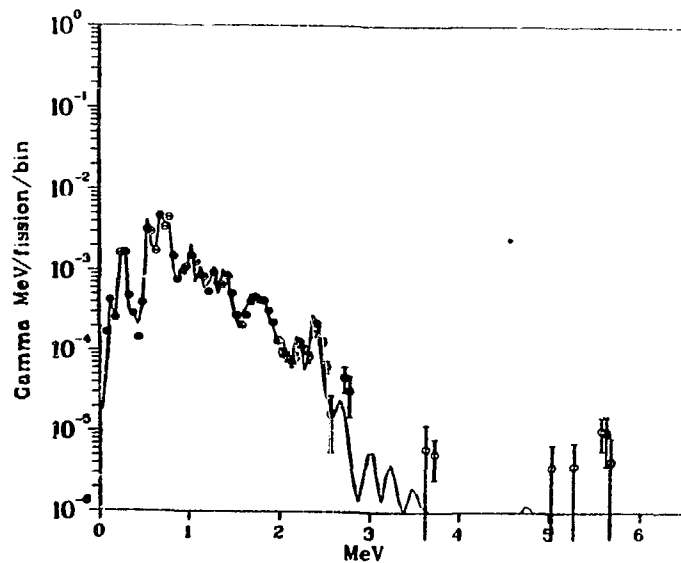
Fig. 13. ^{233}U, after 59 320 s cooling.
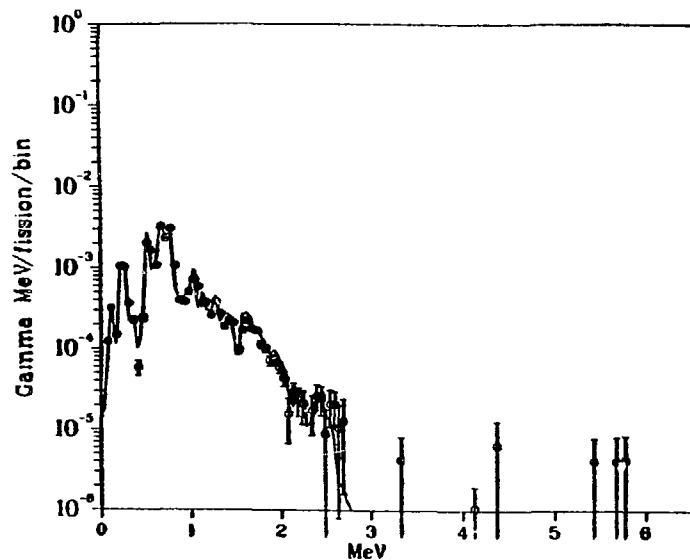
Fig. 14. ^{233}U, after 96 840 s cooling.
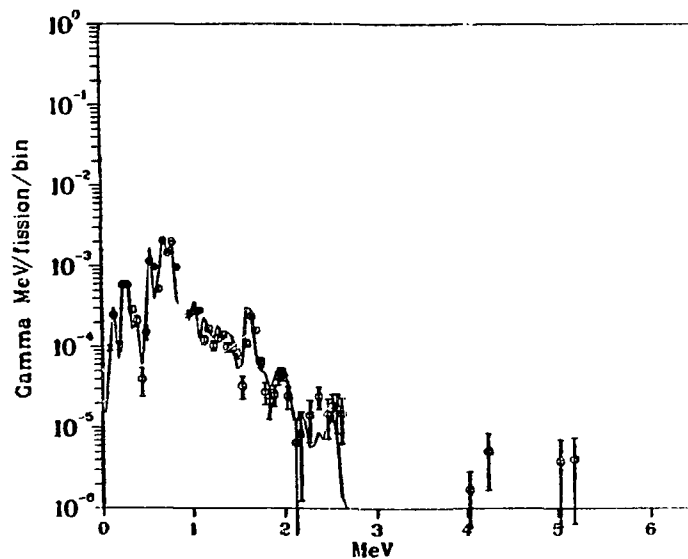
Fig. 15. ^{233}U, after 146 500 s cooling.
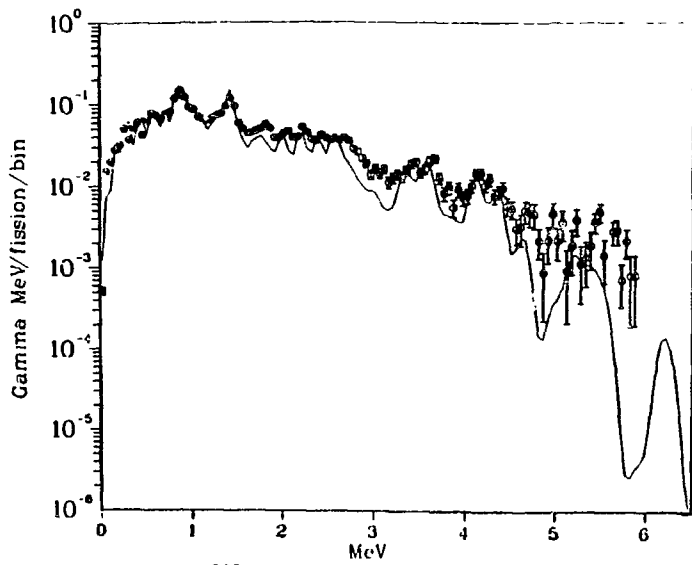
Fig. 16. ^{235}U, after 29 s cooling.
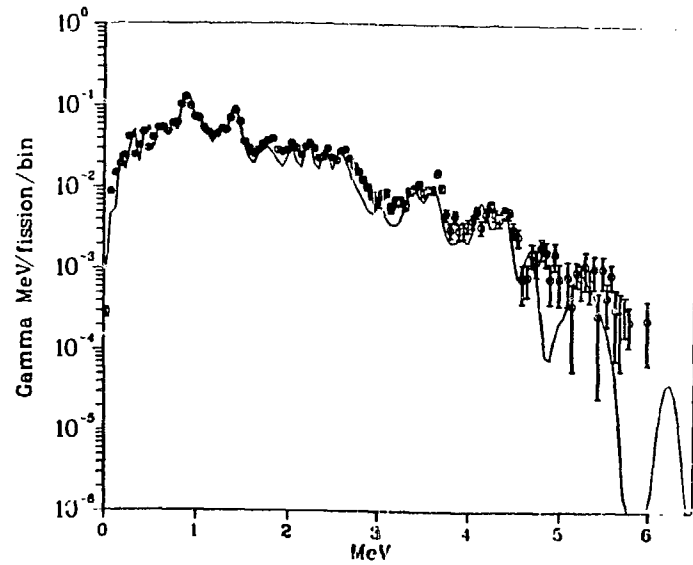
Fig. 17. ^{235}U, after 128 s cooling.
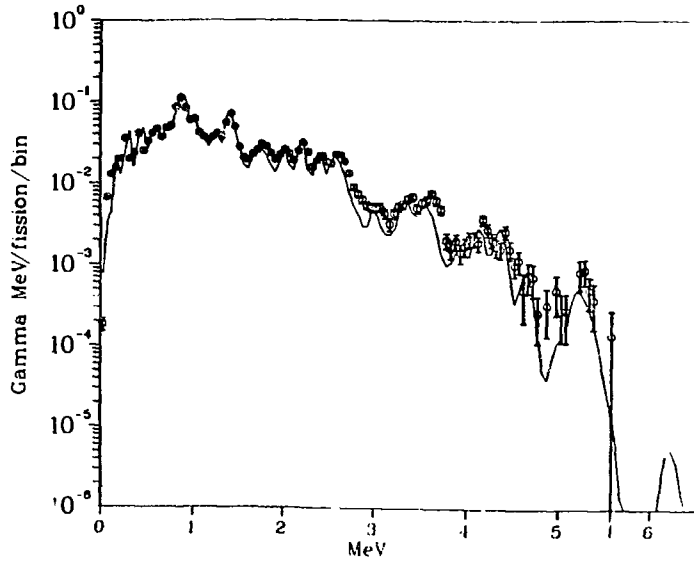
Fig. 18. ^{235}U, after 284 s cooling.
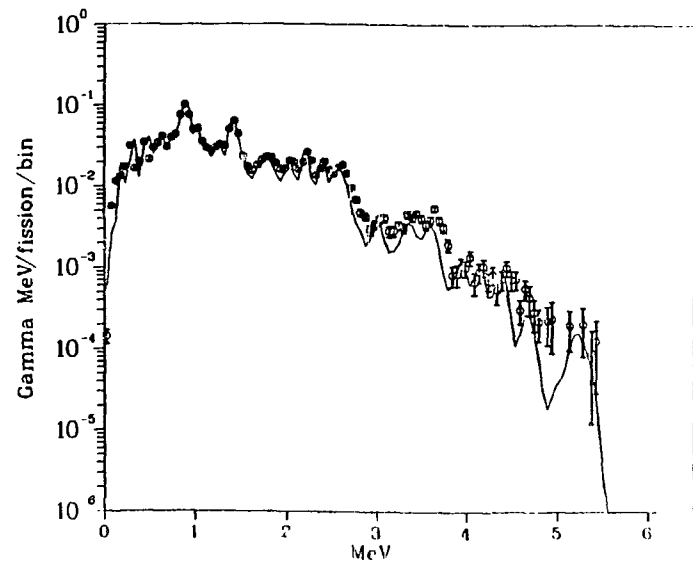
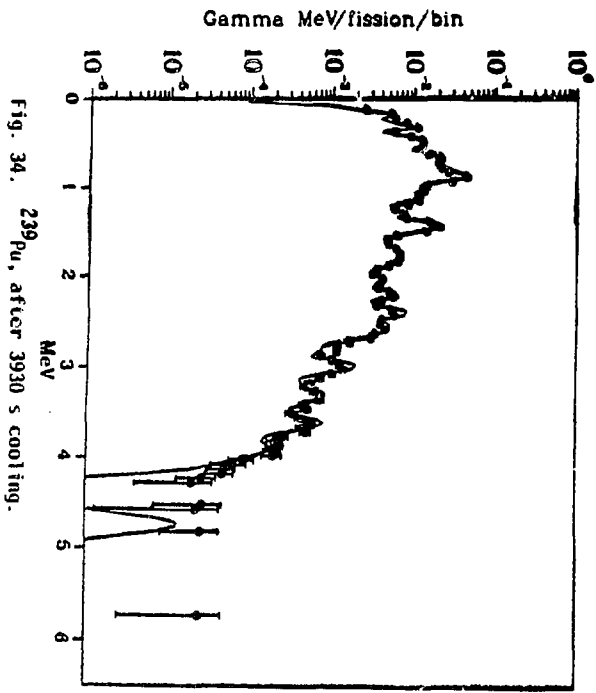
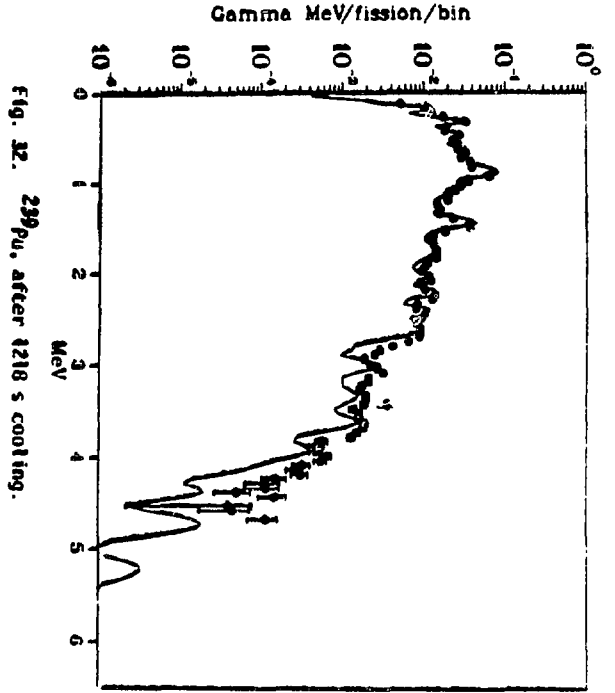Fig. 19. ^{235}U, after 538 s cooling.
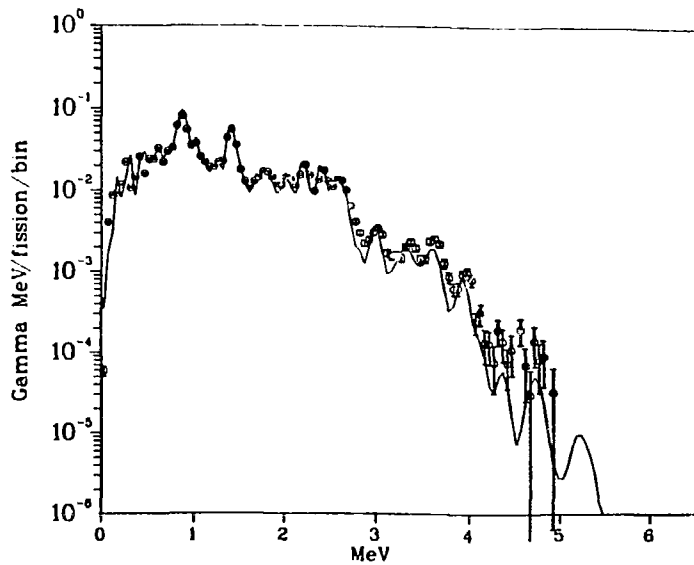
Fig. 20. ^{235}U, after 1218 s cooling.
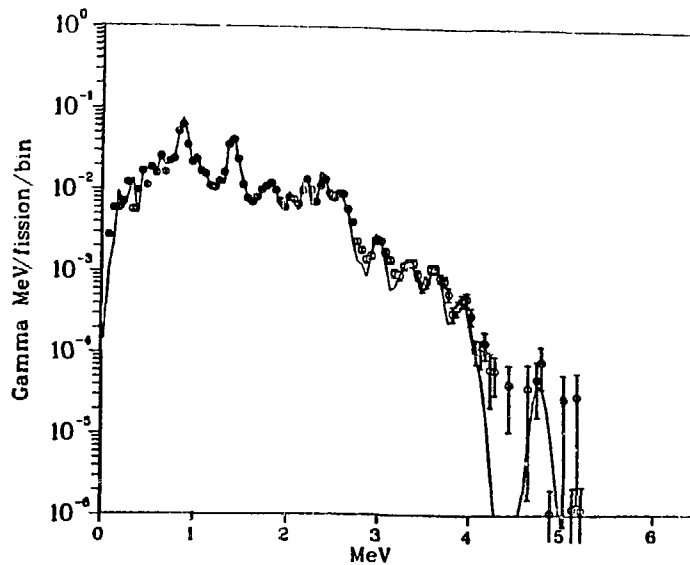
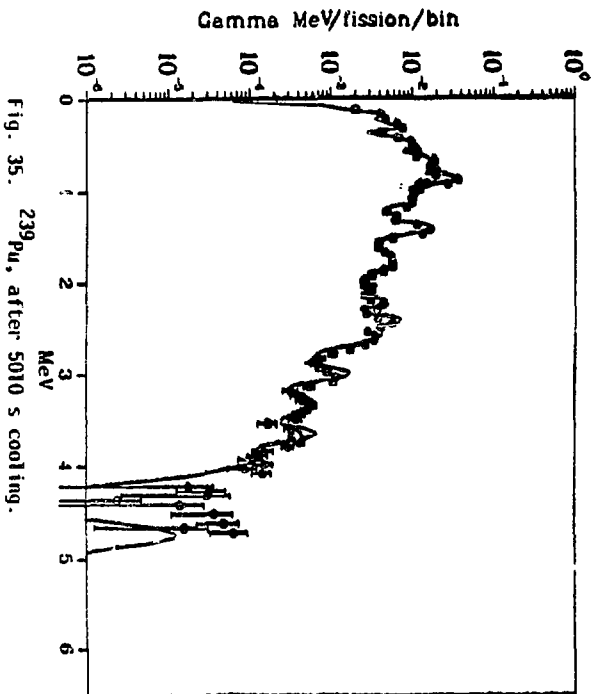Fig. 21. ^{235}U, after 2530 s cooling.
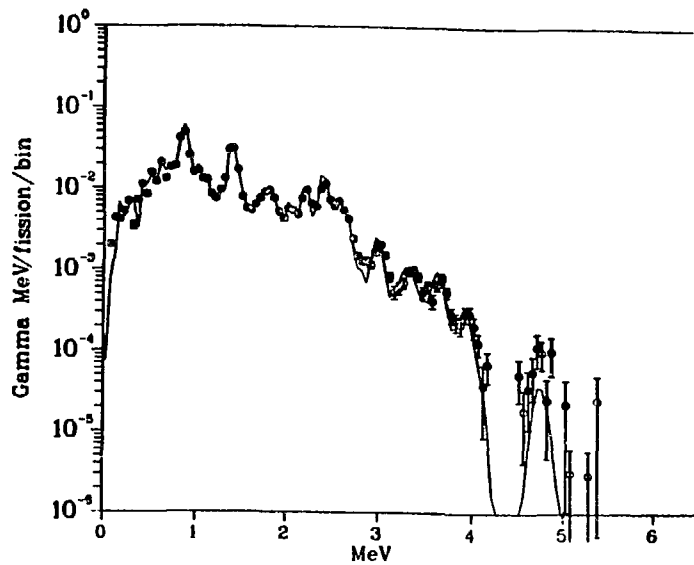
Fig. 22. ^{235}U, after 3930 s cooling.
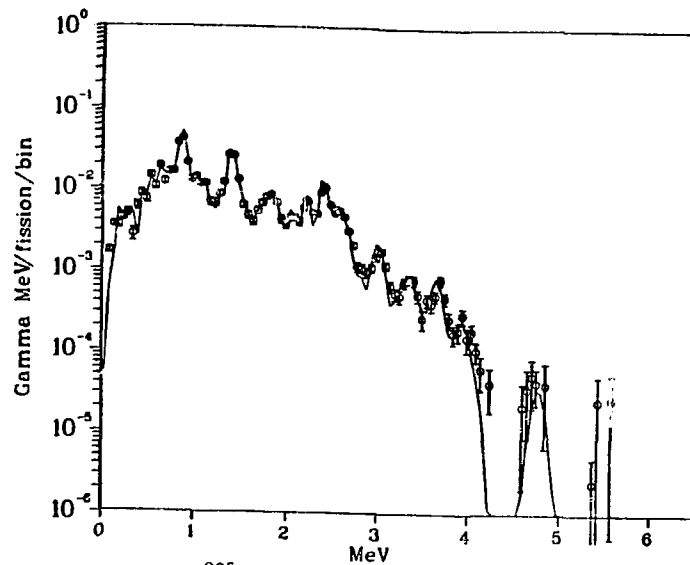
Fig. 23. ^{235}U, after 5010 s cooling.
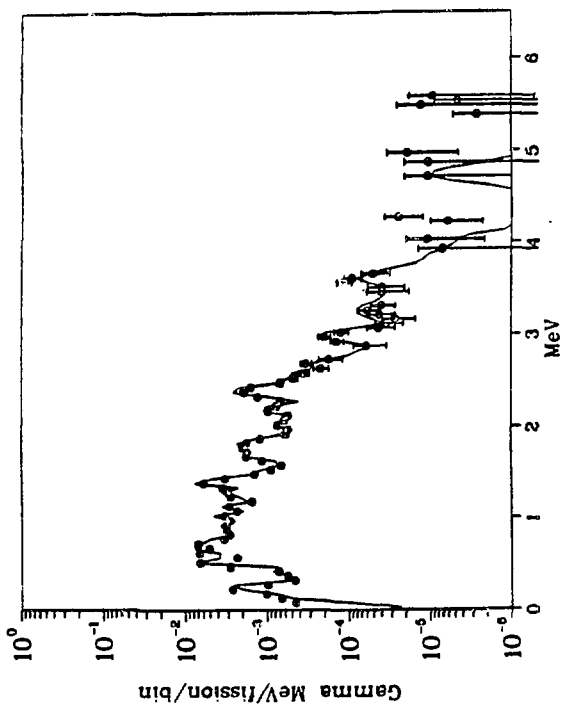
Fig. 24. ^{235}U, after 23 760 s cooling.

Fig. 25. ^{235}U, after 59320 s cooling.

Fig. 26. ^{235}U, after 96840 s cooling.
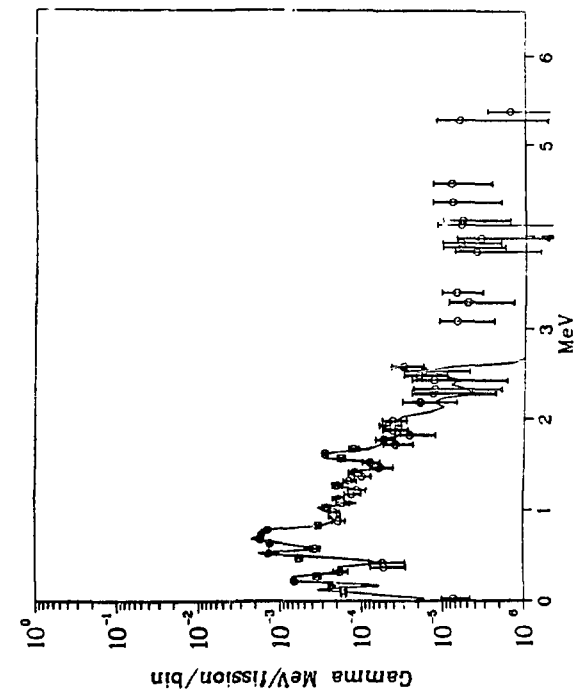
Fig. 27. ^{235}U, after 146 500 s cooling.

17
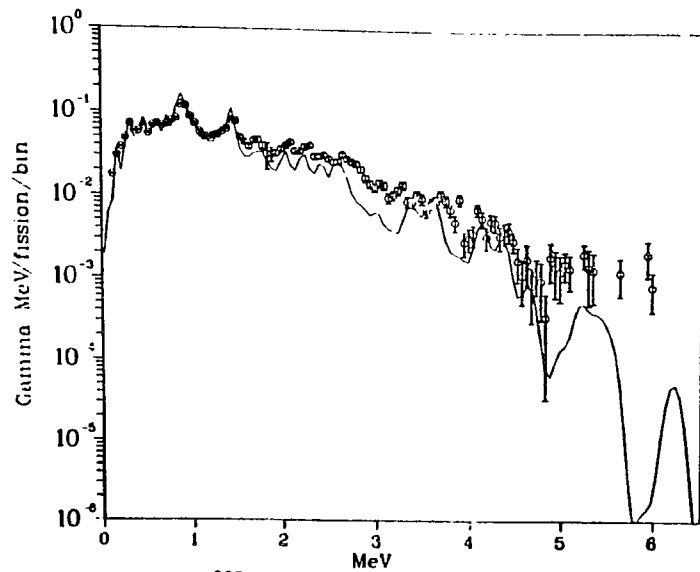
Fig. 28. ^{239}Pu, after 29 s cooling.

Fig. 29. ^{239}Pu, after 128 s cooling.
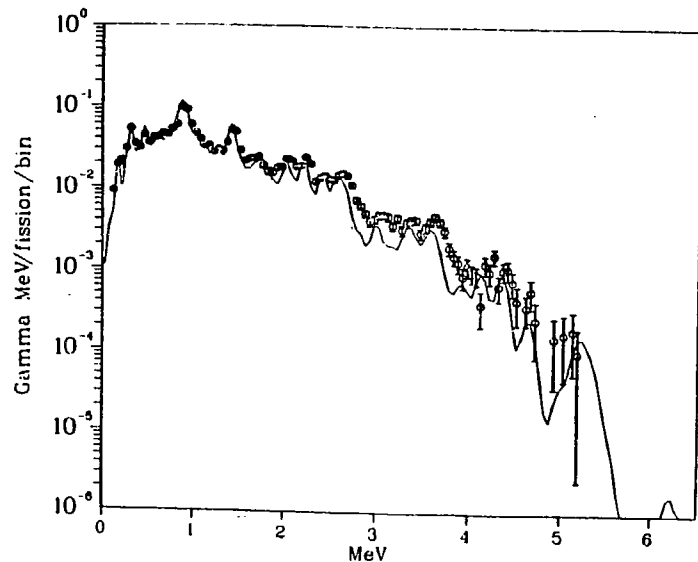
Fig. 30. ^{239}Pu, after 284 s cooling.
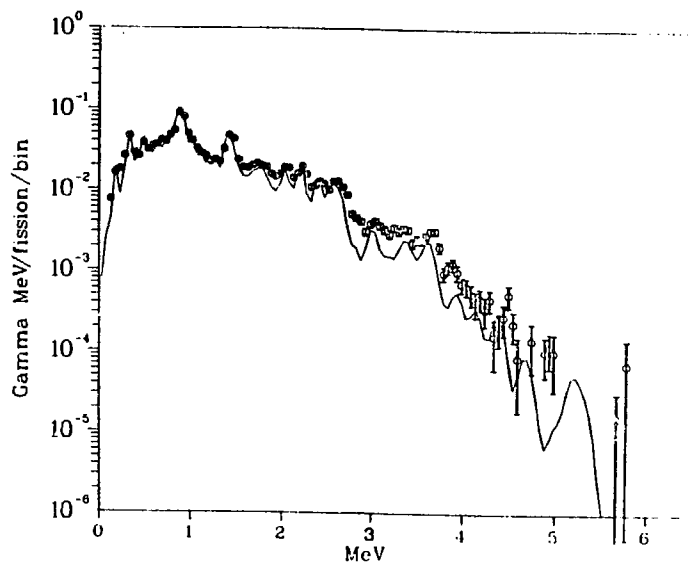
Fig. 31. ^{239}Pu, after 538 s cooling.
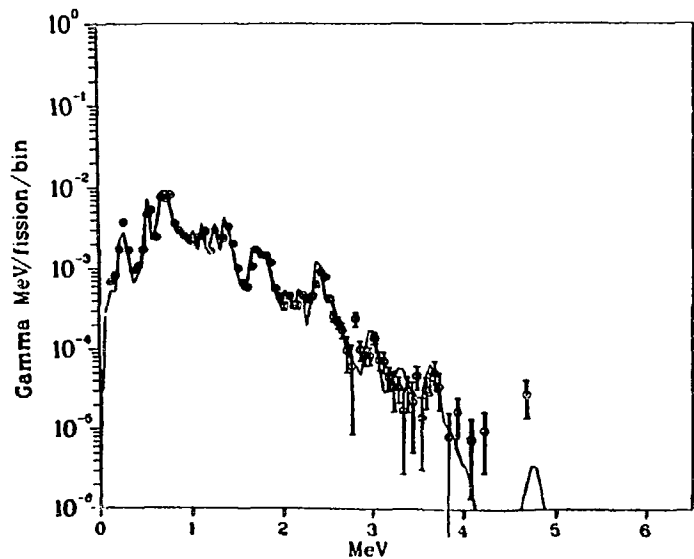
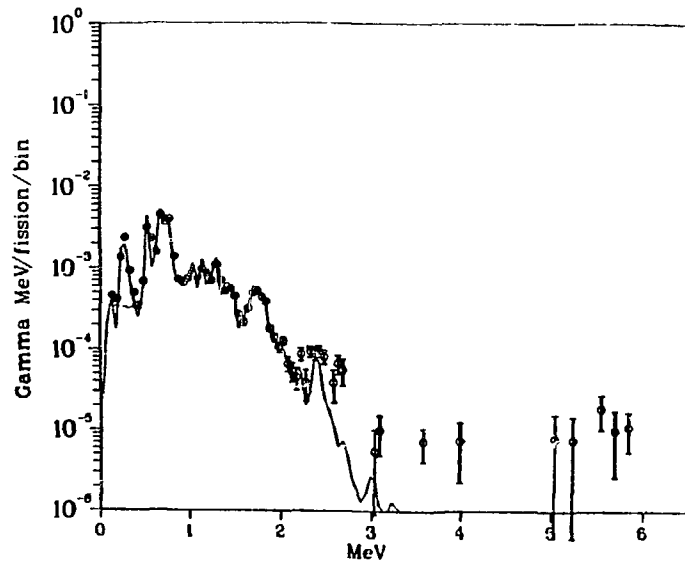Fig. 32. ^{239}Pu, after 1218 s cooling.

Fig. 33. ^{239}Pu, after 2530 s cooling.
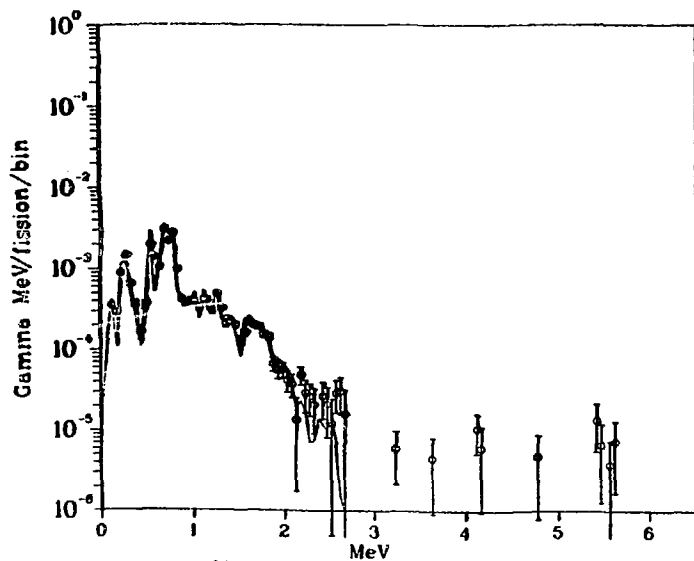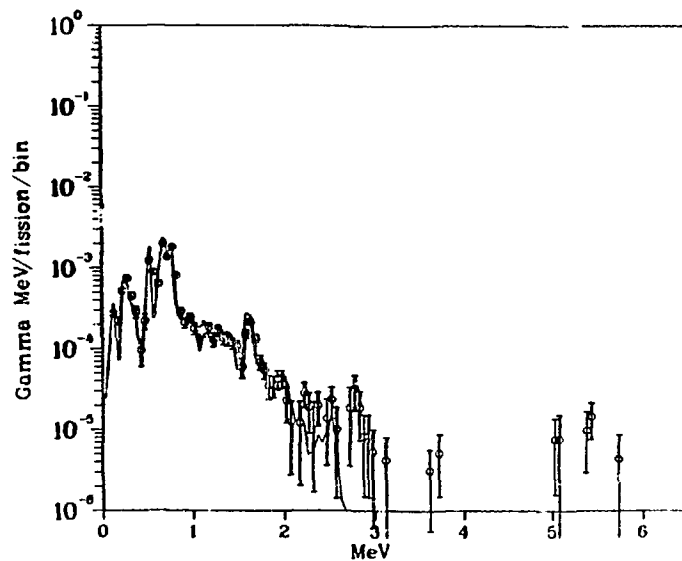

Fig. 34. ^{239}Pu, after 3930 s cooling.

Fig. 35. ^{239}Pu, after 5010 s cooling.

Fig. 36. ^{239}Pu, after 23 760 s cooling.

Fig. 37. ^{239}Pu, after 59 320 s cooling.

Fig. 38. ^{239}Pu, after 96 840 s cooling.

Fig. 39. ^{239}Pu, after 146 500 s cooling.