Decay Heat Measurement of Actinides at YAYOI

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Actinides decay heat was measured for fast neutron fissions in the fast neutron source reactor "YAYOI" of the University of Tokyo. Measured nuclides were U-235 and Np-237, using beta and gamma spectroscopic method. The latest results on beta-ray and gamma-ray decay heat are reviewed in this paper. Measurement results were compared with those measured for U-235 in 1983 by Akiyama using the same method, as well as with the summation calculation results using JNDC-V2, ENDF-B/VI and JEF-2.2.

KEYWORDS:YAYOI, decay heat, uranium 235, neptunium 237, gamma-ray, beta-ray, JNDC-V2, ENDF-B/VI, JEF-2.2

I. Introduction

The decay heat evaluation is important in designing the heat removal system of nuclear reactors and spent fuel handling systems. The actinide decay heat for fast neutron fissions had been measured on U-233, U-235, U-238, Pu-239, Th-232 by Akiyama.[1-4] However, same kind of measurement has not been performed for minor actinides. In this study, minor actinide decay heat was measured using the fast neutron source reactor "YAYOI".[5] Experimental procedures are almost same as Akiyama's one. The decay heat of U-235 was measured covering times following irradiation from 19 to 20,000 seconds, and Np-237 was measured covering times following irradiation from 64 to 20,200 seconds.

II. Experiments

1. Samples

The U-235 samples consisted of about 1.6mg of the metallic fissile materials electrodeposited on 18mm diameter, 0.1mm thick titanium foils. The diameter of the electrodeposited area was 10mm. The Np-237 samples consisted of about 0.5mg of the nitride fissile materials electrodeposited on 18mm diameter, 0.1mm thick titanium foils. The diameter of the electrodeposited area was 10mm. The enrichment is 97.652% for U-235, and 99.9% for Np-237. Each sample was covered with thin myler film, and packed in a thin polyethylene sack. Dummy samples were prepared, which were the same configuration as the U-235 and Np-237 sample except that the fissile materials were not electrodeposited on the titanium foils. The dummy samples were used to estimate the activity of the titanium foil.

2. Irradiation

The samples were irradiated at the center of the grazing hole at YAYOI for time periods of 10 and 100 seconds for U-235, 100 and 500 seconds for Np-237 as well. The cross-sectional view of YAYOI and the irradiation position are shown in **Fig.1**. The samples were transported by air pressure to the irradiation position and rapidly returned to a counting room following the irradiation.
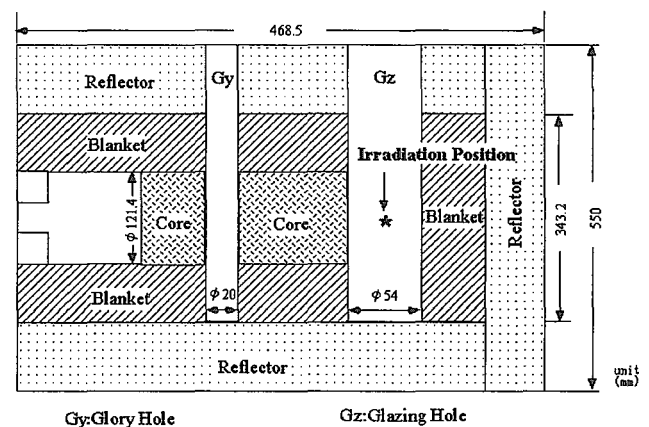

Gy:Glory Hole   Gz:Glazing Hole

Fig.1 Cross-sectional View of YAYOI and Irradiation Position

3. Gamma-ray Detector

Gamma-ray energy spectra were measured by using a NaI(Tl) scintillation detector located inside the lead box. The front face of the detector was covered by 30mm thick polyethylene plate in order to prevent beta-rays from being detected. The distance from the sample to the front face of the detector was 100mm. As this detector had been used by Akiyama, the response function of the detector obtained experimentally by him[1] was used for the data processing.

4. Beta-ray Detector

Beta-ray energy spectra were measured by using a plastic scintillation detector combined with a proportional counter to eliminate gamma-ray effects. The windows were coverd with poly-para-xylene films electrodeposited by gold (total thickness $0.15mg/cm^2$). The distance from the sample to the front face of the detector was 3mm. The proportional counter is filled with a gas mixture, which consists of 90% argon and 10% methane. The response function of the detector

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obtained experimentally by Akiyama[2] was used for the data processing.

Conceptual view of the decay heat measurement system is shown in **Fig.2**.
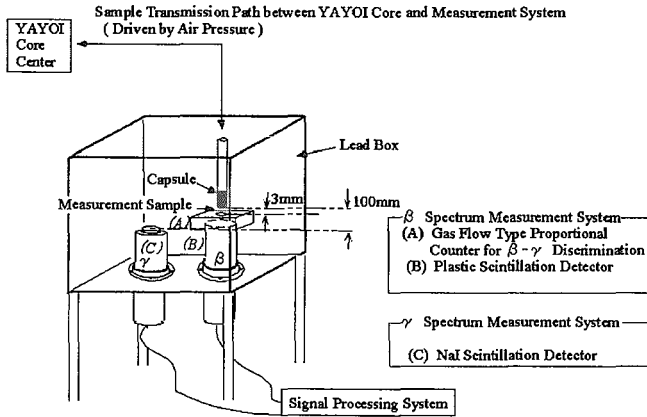

Fig.2 Conceptual View of Decay Heat Measurement System

5. Estimation of Fission Number

The fission number was evaluated from measured gamma spectra by Ge detector, in order to normalize the decay heat measurement data. The decay series to be used for evaluating the number of fission should have a large fission yield, sufficient gamma-ray intensity, and nuclides with proper gamma-ray energy and half life. Nb-97, Nb-97m, Y-91m, and Xe-135 were used to evaluate the number of fission. Decay schemes of these nuclides can be approximated by a simple decay series. For example, in a decay chain of mass number 97, the preceding nuclides of the Zr-97 have rather short half life compared with that of Zr-97. And the independent fission yields of Nb-97m and Nb-97 are negligible compared with the cumulative yield of Zr-97. In this case, the decay chain is simplified as follows:

$$\text{Zr-97} \rightarrow \text{Nb-97 (Nb-97m)} \rightarrow \text{Mo-97}$$
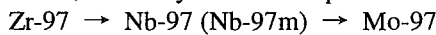
III. Analysis

Measured pulse height data were corrected for background data, and unfolded by using the FERDO[6] code with the use of the response function of the detector. Each unfolded spectrum was divided by the number of fission per second to obtain the normalized spectra. And, the finite irradiation decay heat data were obtained from the normalized spectra.

The finite irradiation decay heat was directly obtained by the present experiment. But, it can not be compared with other experimental results and calculation results obtained under different irradiation conditions. So, it is convenient to convert the finite irradiation decay heat into the fission burst decay heat. If

$$T_W \gg T_R + T_C \cdot \cdot \cdot \cdot \cdot \text{(1)}$$

T_W : waiting time
T_R : irradiation time
T_C : measurement time

is established, the fission burst decay heat is as follows:

$$f(t) = F(t)/T_R \cdot \cdot \cdot \cdot \cdot \text{(2)}$$

f(t) : fission burst decay heat
F(t) : finite irradiation decay heat

If the condition of equation (1) is not established, the correction factor is required. The correction factor is as follows:

$$\eta = f_{cal}(t)/(F_{cal}(t)/T_R) \cdot \cdot \cdot \cdot \cdot \text{(3)}$$

η : correction factor,
$f_{cal}(t)$: result of the summation calculation based on a condition of the fission burst decay heat
$F_{cal}(t)$: result of the summation calculation based on a condition of the finite irradiation decay heat

IV. Experimental Results
1. Results on Gamma Decay Heat for U-235

The experimental result on gamma decay heat for U-235 is shown in **Fig.3**. The present results on U-235 are compared with the data of Akiyama that were measured in 1983 using the same method and summation calculations[7] results based on JNDC-V2, ENDF-B/VI, and JEF-2.2. The present results agreed with Akiyama's data within experimental error. This agreement suggests the repeatability of Akiyama's method. And, the present results agree with a summation calculation results using JNDC-V2 and ENDF-B/VI.
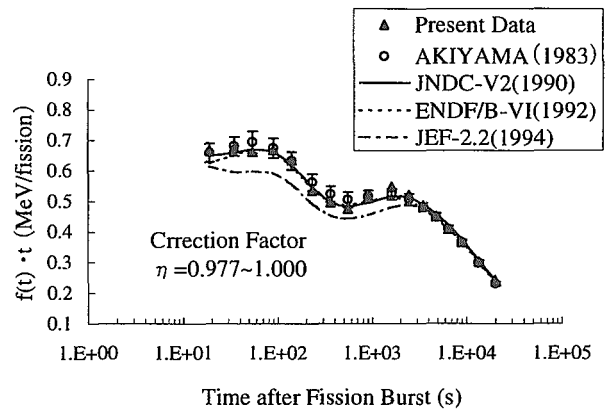

Fig.3 Gamma Fission Burst Decay Heat for U-235

2. Results on Gamma Decay Heat for Np-237

The experimental result on gamma decay heat for Np-237 is shown in **Fig.4**. The present results on Np-237 are compared with the summation calculations results based on JNDC-V2, ENDF-B/VI, and JEF-2.2. The present results on Np-237 agree with a summation calculation results using JNDC-V2 and ENDF-B/VI between 200 and 2,500 seconds. Between 60 and 200 seconds the agreement is not so well. It may be caused by more significant correction factor needed

in this time period. To improve accuracy of decay heat data in early stage after fission burst, another experiment by reducing the irradiation may be useful. Agreement between the present results and the analysis results is bad between 2,500 and 20,000 seconds. In this time period, gamma-rays released from Np-238, that were generated by neutron capture reaction of Np-237, can not be ignored. **Figure.5** shows gamma spectrum from irradiated Np-237 measured by Ge detector about 56,000 seconds after irradiation. Four gamma-ray peaks were seen. These peaks were not seen on U-235 irradiation data, titanium foil irradiation data, and background data, at all. Main gamma energy and release rate of Np-238 are 923.98keV(2.86%),984.45keV(27.8%),1025.87keV(9.65%), and 1028.54keV(20.385%). Observed four gamma-ray peaks agree with each energy, respectively. And, the ratio of the peak count rates are consistent with the gamma release rates. It is also confirmed that the transition of these peak counts can be explained by the half life of Np-238, 2.12 day. As for the time period between 2,500 and 20,000 seconds, the decay heat corrected for four gamma-ray peaks of Np-238 agreed with the summation calculation result using JNDC-V2. **Figure.6** shows improvement of the elimination of Np-238.
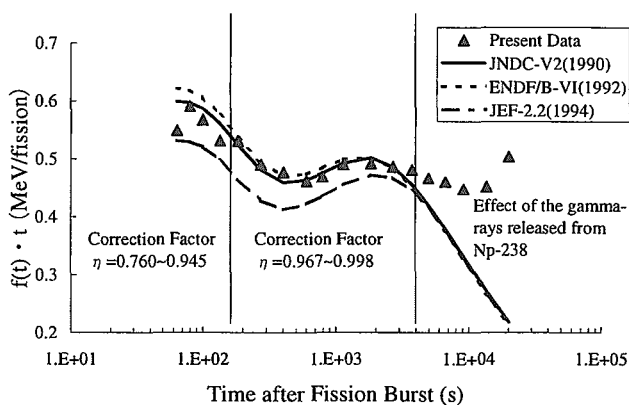

Fig.6 Np-237 Gamma Decay Heat Corrected for Gamma-rays of Np-238
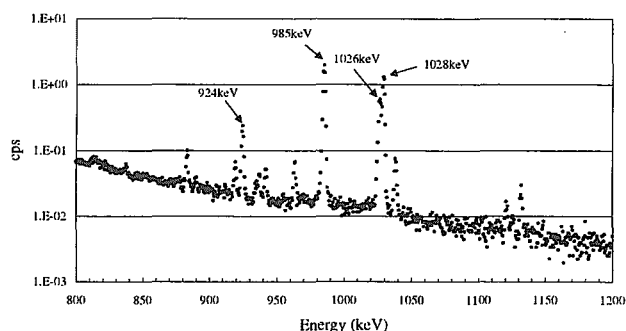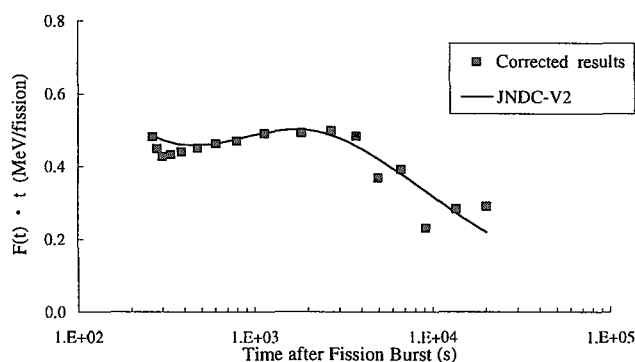
3. Results on Beta Decay Heat for U-235 and Np-237

The experimental result on beta decay heat for U-235 is shown in **Fig.7**. The present results on U-235 are compared with the summation calculations results using JNDC-V2. The present results on U-235 agreed with the summation calculations results using JNDC-V2 between 140 and 230 seconds. However, between 360 and 13,500 seconds, the present results were smaller than the summation calculations results using JNDC-V2. Calculated to experimental (C/E) value was 2.12 in maximum.

Beta decay heat for Np-237 was also measured by using the same method for U-235. The large discrepancy was also found in the Np-237 data, as well.
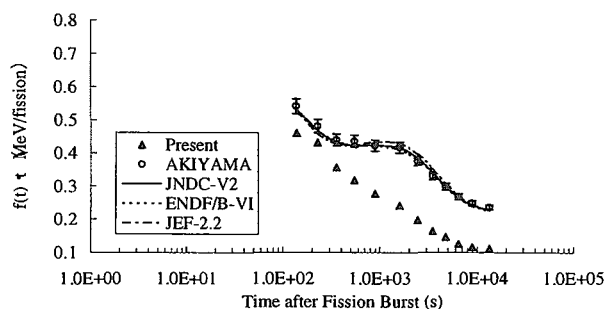

Fig. 4 Gamma Fission Burst Decay Heat for Np-237

Fig.7 Beta Fission Burst Decay Heat for U-235

Beta-ray energy range available in the present measurement was between 0.3 and 4MeV. Fraction of beta decay heat for energy region 0.5-4MeV is 88%, so main energy region was covered by the present measurement. Energy distribution of beta-ray for U-235 is shown in **Fig.8**. In early time period after fission burst, the experiment results agreed with the calculations results. However, in late time period after fission burst, a large difference was found between the experiment results and the calculations results. Same tendency was confirmed in the Np-237 data, as well.

The response function of the beta-ray detector can be considered to one of the reasons of a large difference. Then, the measured value of a beta-ray of Sr-90+Y-90 was compared with the calculation value. Comparison between measured and calculated beta-ray spectrum is shown in **Fig.9**.

Fig.5 Gamma Spectrum of Irradiated Np-237 by Ge Detector (about 56,000 seconds after irradiation)

The ratio of measured spectrum and the calculation spectrum was made into the correction factor. When the present result of U-235 and Np-237 was multiplied by the correction factor, it was improved by increasing of about 15%. However, there was no change in C/E value decreasing in connection with cooling time.

Considering the above-mentioned information, the present beta decay heat could not be considered as reliable as those obtained in the gamma decay heat measurement.
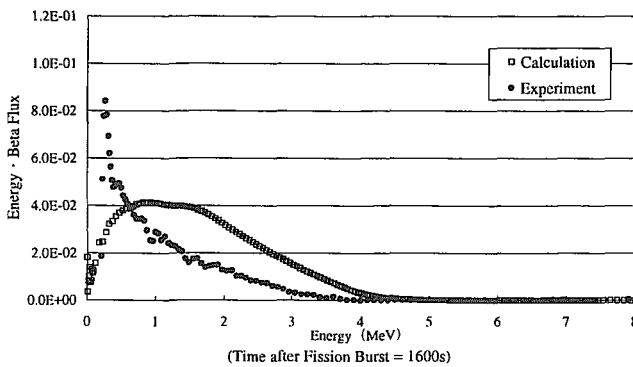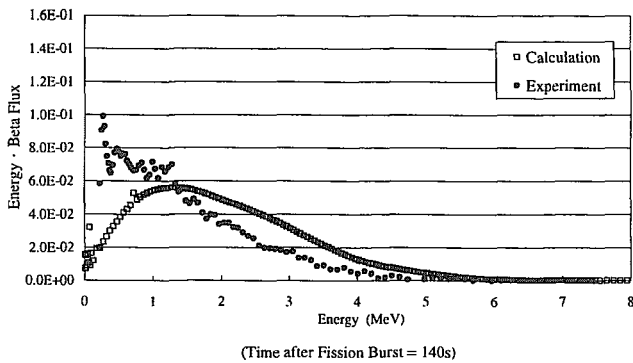

(Time after Fission Burst = 140s)

(Time after Fission Burst = 1600s)

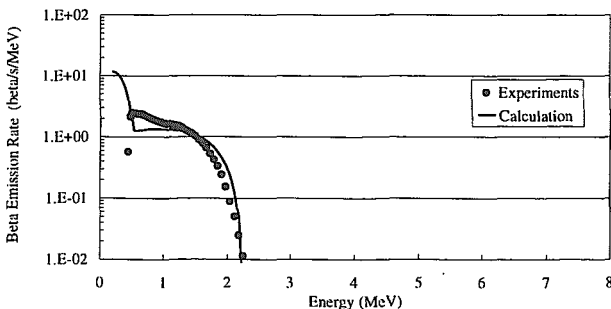**Fig.8** Energy Distribution of Beta Ray for U-235

Fig.9 Comparison between Measured and Calculated Sr-90+Y-90 Beta-ray Spectrum

V. Conclusion

The decay heat of U-235 for fast neutron fission was measured covering times following irradiation from 19 to 20,000 seconds, and that of Np-237 was measured covering times following irradiation from 64 to 20,200 seconds.

Experimental method is same as Akiyama's one by which the decay heat of U-235, Pu-239, etc, had been measured.

The present results on gamma decay heat for U-235 agreed with Akiyama's data within experimental error. And, the present results agree with a summation calculation results using JNDC-V2 and ENDF-B/VI. The present results on gamma decay heat for Np-237 agree with a summation calculation results using JNDC-V2 and ENDF-B/VI between 200 and 2,500 seconds. Between 60 and 200 seconds, the agreement is not so well. The reason may be more significant correction factor needed in this time period. As for the time period between 2,500 and 20,000 seconds, the decay heat corrected for four gamma-ray peaks of Np-238 agreed with a summation calculation result using JNDC-V2.

Beta decay heat was also measured by using the same method for gamma decay heat. In early time period after fission burst, the present results on beta decay heat for U-235 and Np-237 agreed with the summation calculations results using JNDC-V2. On the other hand, in late time period after fission burst, the present results were smaller than the summation calculations results using JNDC-V2. In comparison of energy distribution of beta-ray for U-235 and Np-237, a large difference was found between the experiment results and the calculations results. However, the reason for a large changes in C/E value is not confirmed.

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