Detection of gamma ray from short-lived fission product at KUCA and KURNS-LINAC

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Two kinds of γ ray spectrum measurement are performed. The one is for a critical core of enriched uranium (U) mocked up in Kyoto university critical assembly facility (KUCA). The other is for a U sample of natural enrichment to obtain microscopic nuclear data at an accelerator neutron source facility (KURNS-LINAC). In KUCA, discrete energy γ rays from 2.9 to 5 MeV are observed above prompt components. In KURNS-LINAC, also discrete energy γ rays of resemble spectrum are measured in a time-background region. By comparing the measured spectra to the numerical analyses of production and decay of isotopes originated in fission of ^{235}U, those discrete gamma rays are identified as those from decay of short -lived fission products.

1. Introduction

Fission products (FPs) play significant roles in power reactors. For example, the delayed neutron emissions from FPs mitigate power transient due to a reactivity insertion and the decay heat of FP may initiate a severe accident if it is not removed due to the station black out. In these regards, production and decay of FP must be predicted accurately. For those purposes, the data of the yield and the decay chains of FPs have been evaluated and compiled in libraries such as JENDL/FPY & FPD-2011 [1]. Those libraries have been developed based on γ ray spectra measured with scintillators and validated against measured decay heat [2,3]. However, they are seldom to be validated against isotopic data since γ ray data from short-lived FP are scarce.

Two kinds of experiments were conducted in the Kyoto university critical assembly facility (KUCA) and the LINAC neutron source facility of the institute of integrated radiation and nuclear science, Kyoto university (KURNS). In KUCA, γ ray spectrum was measured with a highly purified germanium (HP-Ge) detector for a critical core [4,5]. In KURNS-LINAC, U metal sample of the natural enrichment was irradiated by neutrons of white spectrum and the emitted γ rays from the sample were measured with the same HP-Ge [6]. In the two experiments, resemble γ ray spectra were measured unexpectedly. In this work, the measurement and numerical analyses for the discrete γ rays are discussed to see whether the measured data are feasible for the validation of the yield and the decay chains of FPs.

2. Critical experiment

Details of the critical experiments and the identification of FPs had already been presented [4, 5], so only essential points are described. Schematic view of the set up is shown in Figure 1. The critical core was mocked up with fuel plates of uranium (U) - aluminum (Al) alloy, of which ^{235}U enrichment is 93 wt-%. The criticality is adjusted by the control rod position. 428 sheets of the plates were loaded into the C-core tank of KUCA. The effective core size was about 28 cm x 38 cm x 57cm. The fuel plates were immersed in the light water. The core power was adjusted less than 4.6 mW. Outside the tank, a HP-Ge detector of the 35 % relative efficiency was put. Between the fuel plates and HP-Ge, there was light water layer of 60 cm thickness. By the arrangement, the neutron flux was well attenuated in the light water and the transmission γ rays were measured. γ ray count rate was 13 kcps.

γ ray pulse height spectra of good resolution were obtained [4, 5]. In the spectra, 2.223 and 7.724 MeV γ rays from radiative capture of hydrogen (^{1}H) and ^{27}Al, respectively, are prominent. The spectra in energy region from 2.9 to 3.9MeV are magnified in Figure 2. The count rate is mainly given by the continuum spectrum due to the fission prompt γ rays.

Above the continuum spectrum, discrete energy γ rays are found. Distinct peaks are of the photo-electric, the single escape, and the double-escape components of γ rays from ^{27}Al(n,γ) reactions. However, peaks of fewer count rates are observed. By comparison of the small peaks to the calculation described in Section 4, the peaks are considered to be the components due to short lived FPs such as ^{87,88}Br, ^{89}Kr, ^{90,90m,91}Rb, ^{95,97,98}Y, ^{136}Te. Since 93 wt% -^{235}U metal fuel was used in the water, almost all the FP were originated in fission of ^{235}U. As for ^{90}Rb, the γ ray peak of 4136 keV is overwrapped by ^{27}Al(n,γ) components. However, they were confirmed by the measurement posterior to the critical operation. It was done for three minutes after the control rod drop. However, γ rays of half-life shorter than 1 minutes could not be measured posterior to that due to the statistical fluctuation. It is the advanced feature of the measurement under the critical condition to enable detections of γ rays from FPs of half-life shorter than 1 min (^{97,98}Y, for example). Whereas, due to the prompt components, the peak to base ratio was small.

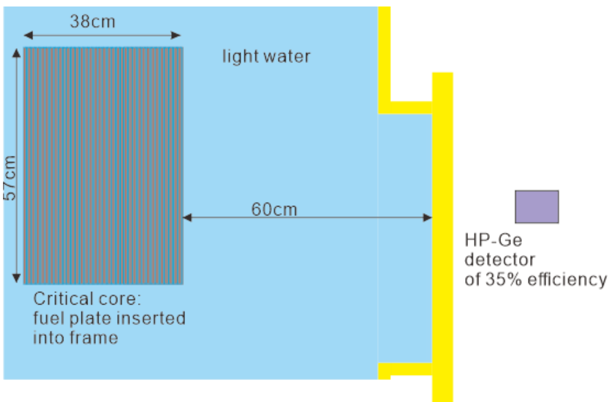

Figure 1. Side view of the critical core and HP-Ge detector. Yellow lines are of aluminum alloy.

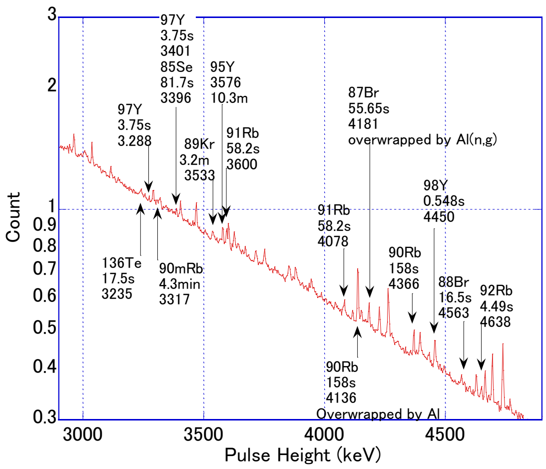

Figure 2. Measured γ ray spectrum for critical core.

3. Microscopic Nuclear Data Measurement

The original purpose of the microscopic nuclear data measurement was to obtain γ ray spectrum from reactions of ^{238}U induced by neutrons of thermal and resonance energies. The energy of the incident neutron is determined by the time of flight (TOF) of the neutron between the target and the sample.

At KURNS-LINAC facility, accelerated electrons were injected onto a tungsten (W) target and bremsstrahlung rays were radiated. Then photo-nuclear reactions were induced, and fast neutrons were emitted. The neutrons were moderated to the resonance and thermal energy regions in the light water surrounding the W target. U metal sample was set effectively 11.3 m from the W target. The size of the sample was 4 cm x 4 cm and the thickness was 2.91 g/cm^2. For the TOF measurement, pulsed neutron source was used with frequency of 50 Hz. The same HP-Ge was set 5 cm from the sample. The detector was shadowed from the neutron and the γ ray flash from the W target with a collimator. The energy dependent γ ray detection efficiency was measured by detection of the γ ray spectrum from the radiative capture of ^{35}Cl.

The TOF spectrum was shown in Figure 3. Events of the earlier TOF is generally induced by the faster neutrons. The events induced by fast neutrons of energy greater than keV could not be measured since influence by the γ ray flash from the W target still remains in earlier TOF. The peak spectrum in Figure 3 corresponds to the radiative capture at the resonance neutron energy. In the lower energy, broad peak structure is found. It is called "thermal peak". After the decay out of the thermal peak, flat component is found, which is treated as the time background components for the original purpose. In the present work, the γ ray pulse height spectrum in the time background region was focused on. Figure 4 shows the pulse height spectrum. Since the time region was after the thermal neutron decays out, prompt γ rays from radiative captures and the fissions do not exist. Accordingly, this component was considered due to decay of FP of half-life longer than 10 ms. By comparison of the peaks to the data in Figure 2, the same γ rays were measured although ^{92}Rb, ^{88}Br, and ^{136}Te are not observed. Generally, the counting statistics is poorer in this time-background condition. Instead, the better peak to base ratio was given thanks to the attenuation of the prompt γ rays from the radiative capture and the fission reactions.

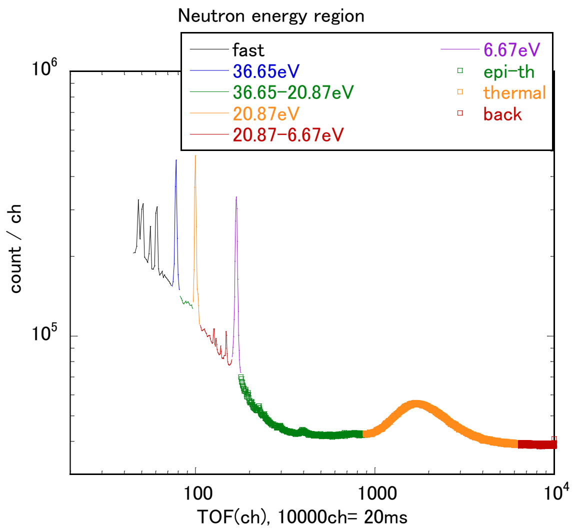

Figure 3. Time of flight spectrum for estimation of energy of incident neutron.

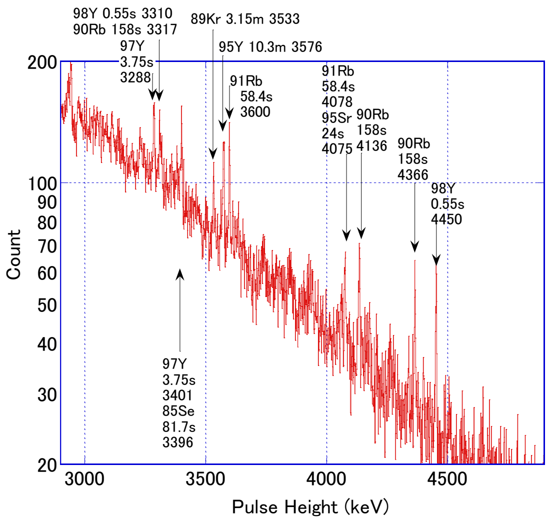

Figure 4. γ ray spectra at time background. Nuclides in labels denote β-decay parents of delayed γ rays.

0.72% of the metal sample was ^{235}U and that was also irradiated by the thermal neutron. Accordingly, a part or almost all FPs might be generated by fission of ^{235}U. However, it should be also noted again that neutron flux in energy region higher than keV had never been characterized due to the γ ray flash. Accordingly, there is a possibility that some FPs are generated by fission of ^{238}U induced by neutrons faster than the threshold energy (~MeV).

4. Comparison to Calculation

In the both measurements in KUCA and KURNS-LINAC, the neutron flux was low so that radiative captures of produced FPs can be neglected. In such cases, the activities of FPs are determined by the radioactive decay and the fission rate. In this work, it was assumed that all FPs were produced by fission of ^{235}U induced by the thermal neutrons. The depletion after the instant irradiation was calculated by Bateman's method [7] in an exact manner, based on the JENDL/FPY & FPD-2011 library. Then the activity was integrated for time lengths of the neutron irradiation and of the γ ray measurement. For the critical experiments, the flux variation with time was also considered. For the microscopic data measurement, fission rate was assumed as the continuous wave operation and the measurement time was also continuous, in spite that the fission occurred periodically induced by the pulsed neutron source and only γ ray spectrum during the time background region was measured. Then using the data in JENDL/FPD-2011, the number of emissions of the discrete γ rays were deduced. The measured γ rays were identified by comparison to the calculated γ rays of larger emission rates as shown in Figures 2 and 4.

Quantitative analyses were also performed. For the critical experiments, the γ ray pulse height spectrum was simulated with the MCNP-5 code [8] and the AcelibJ40 library [9] based on JENDL-4.0. At first, the fission reaction distribution in each fuel plate was estimated by a neutron transport calculation in the manner of the eigenvalue calculation. In the next step, the γ rays of each energy shown in Figure 5 were transported from the fuel. The spatial distribution of the γ ray emission is treated similar to that of calculated fission reaction rates. In the calculation, the threshold energy of the γ ray transport was set 1.6MeV outside the HP-Ge detector. Inside the detector, it was set down to 3 keV. For the tally, photon - electron coupled transport was done and the pulse height response was calculated. The calculated count rate for the pulse height was compared to the measured one in Figure 5.

For the microscopic data measurement, the experimental count rates were divided by the energy dependent efficiency measured using the $^{35}Cl(n, \gamma)$ reaction. The deduced ratio is considered proportional to the number of photon emission. Then, the calculated one was compared to the deuced one in Figure 6.

In the both figures, the absolute values of C/E do not make sense since the absolute efficiency was not known in the critical core and the continuous wave operation was assumed in the microscopic data measurements. C/E for each γ ray depends on FP and γ ray energy. However, variation of C/E with FP is resembled each other in the two measurements. That indicates significant part of the discrete energy γ rays were radiated from the same FP produced by the fission reactions of the same fissionable nuclide. Accordingly, the two kinds of experiment are considered worth to be investigated further to obtain the reference data for validating FP yield and decay data libraries.

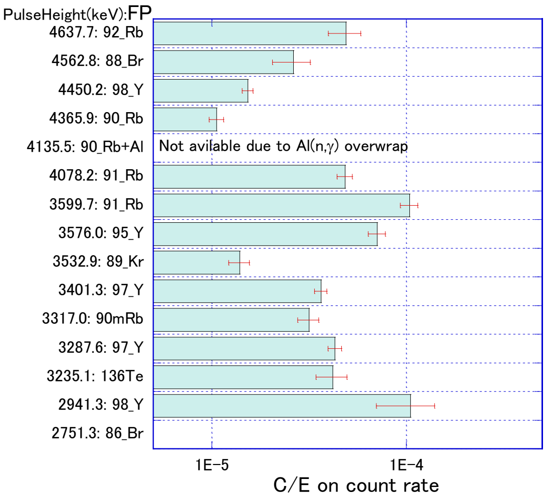

Figure 5. Comparison of calculated photo-electric peak counting to that by the measurement in KUCA.

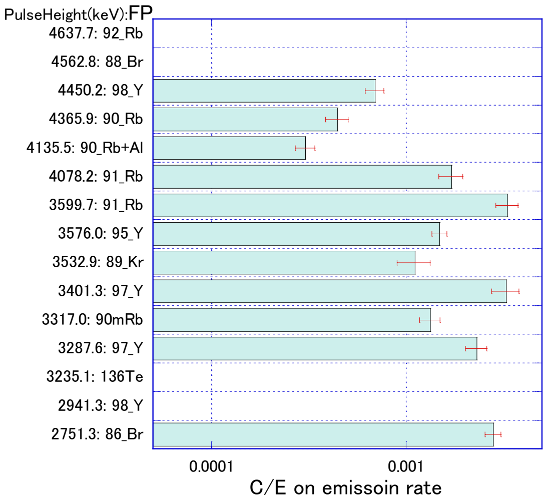

Figure 6. Comparison of calculated number of γ ray emission to that by measurement in KURNS-LINAC.

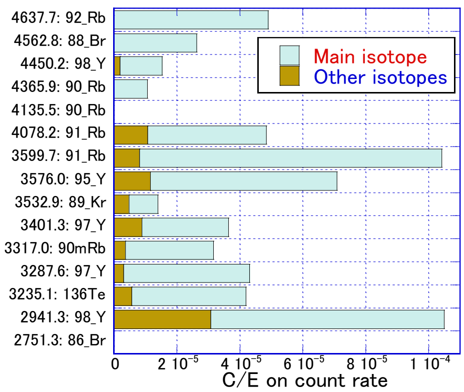

Figure 7 Contribution of other FPs to the count rate of target FP.

For the validation, additional measurements or improvements of the methods are required. For the critical core in KUCA-C, experimental methods to determine the energy dependent γ ray detection efficiency is desired. Besides, analyses of the prompt components by the radiative capture and the fission are mandatory. Whereas, for the microscopic data measurement in KURNS-LINAC, quantification of neutron flux in energy region higher than 500 keV was required to see whether fission reaction of ^{238}U was significant or not. Besides, the numerical analysis of the production of FP in the pulse mode operation and the decay of them in the time region posterior to decay out of the thermal neutrons must be achieved. Another issue is the resolution of the HP-Ge detector. Various energies of γ ray are radiated from FPs produced by the fission of ^{235}U. Consider a measurement with a HP-Ge detector of a resolution of ±5 keV. When we measure γ rays of energy E, γ rays of energy from E – 5 keV to E + 5 keV are measured in the discrete peak. Based on the JENDL/FPY & FPD-2011, the contribution of the main isotope and the other isotopes are calculated. As shown in Figure 7, considerable fraction of counts of a peak are attributed to the other components. In that sense, validation of FP yield and decay data should not be focused only on a single γ ray but comprehensive set of evaluated FPs should be validated against the measured data.

5. Conclusion

At KUCA, γ rays radiated from short-lived FP were measured in a critical core under the intense background of prompt components of fission and radiative capture reactions. At KURNS-LINAC, they were also measured for metallic U sample irradiated by white neutron source in the time region where the thermal neutrons decayed out. The energy of the γ rays and corresponding FPs were identified by comparison of the measurement to the depletion calculations based on JENDL/FPY & FPD-2011 library. γ ray count rates and emission rates were numerically calculated and compared to the measured data of KUCA and KURNS-LINAC, respectively. The ratio of the calculation to the measurement varied with FPs. However, the trend of the ratio for the KUCA experiment is resemble to that for the KURNS-LINAC experiment. The results indicate that the FP γ rays produced by fission reaction of ^{235}U induced by thermal neutrons are the major components in the both measurements. The both measurement techniques are superior in detection of short-lived FPs compered to measurements posterior to neutron irradiation. Accordingly, the γ ray spectroscopy for the critical core and that for a uranium sample posterior to decay - out of the thermal neutrons are promising to give reference data to validate the FP yield and decay data. For that, necessary improvements of the methods shall be conducted.

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