Determination of the Burnup and Pu/U Ratio of PWR Spent Fuel Samples by a Gamma-ray Spectrometry

Kwang-June Park, June-Sik Ju, Hee-Sung Shin, Yong-Bum Chun, Ho-Dong Kim
Korea Atomic Energy Research Institute, P.O.Box105, Yuseong, Daejeon, Korea 305-600
Corresponding author: kjpark@kaeri.re.kr

1. Introduction

Burnup is one of the major factors to identify the safety and economy of nuclear fuel management processes such as a storage, transportation, reuse, and disposal of spent nuclear fuels. Also, the Pu/U ratio is a major factor to account for the nuclear materials in the spent fuels. In this work, a gamma/neutron combined measuring system was newly constructed for a small spent fuel sample application before measuring pyro-processing products. And the burnup and Pu/U ratio of the spent fuels were determined by both a nondestructive and destructive method, that is, a gamma-ray spectrometry and a chemical analysis. And the precision of the burnup and Pu/U ratio results obtained from the gamma-ray spectrometry were evaluated by using the chemical analysis results.

2. Experiment

Gamma-ray measurement of the J502-R13-2 sample was carried out with a newly developed gamma/neutron combined system which is loaded with GENIE-2000 software for a gamma-ray analysis, and the counting time and dead time were 9,000 second and 0.94%, respectively.

3. Results and Discussion

Burnup in atom % can be defined as the ratio of consumed ^{235}U to the total amount of uranium. That is to say, the consumed ^{235}U is the amount of ^{235}U fissioned by the nuclear reaction. Also, the ratio of ^{134}Cs to ^{137}Cs for the amount produced during the reactor operation is proportional to the epi-thermal neutron flux and irradiation time.

In this work, we attempted to determine the burnup and isotope ratio of a spent fuel sample, J502-R13-2, by means of a combined method such as a gamma-ray spectrometry and a computer code calculation.

3.1. Isotope Ratio

In the gamma-ray spectrometry, the isotope ratio is determined from parameters such as the net peak area, relative efficiency, branching ratio, half life, etc. And the isotope ratio in the code calculation depends on the burnup, specific power, initial enrichment and cooling time of the spent fuel to be measured. As a result of the calculation, the value of the $^{134}Cs/^{137}Cs$ ratio was calculated to be 0.0377.

3.2. Burnup-Isotope Ratio Correlation

It is not easy to directly determine a spent fuel burnup by a gamma-ray spectrometry. So, a correlation between a burnup and isotope ratio is needed, it is able to be achieved by a code calculation. The $^{134}Cs/^{137}Cs$ ratios were calculated first with each burnup, and the correlation curve of the burnup-$^{134}Cs/^{137}Cs$ ratio was obtained by fitting the data points as shown in Fig.8. The correlation equation is as follow:

$$Burnup = 5169.82 + 295118.99(IR) + 2.48 \times 10^7 (IR)^2 \quad (1)$$

where IR is the isotope ratio ($^{134}Cs/^{137}Cs$ ratio).

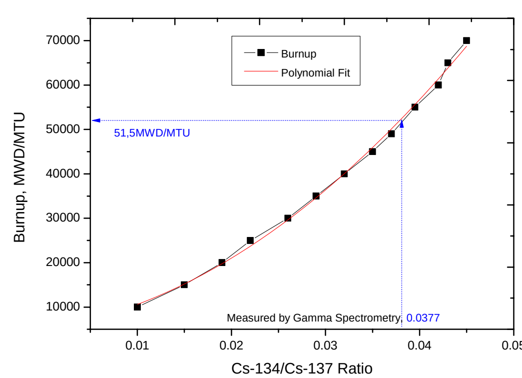

Fig.1. Correlation Curve for the Burnup-$^{134}Cs/^{137}Cs$ Ratio of J502 Spent Fuel.

3.3. Burnp Determination

Burnup of the J502-R132-2 sample was determined by using the $^{134}Cs/^{137}Cs$ ratio measured from the gamma-ray spectrometry and the correlation equation (1) derived from the results of the ORIGEN-ARP code [1] calculation. As a result of inserting 0.0377 ($^{134}Cs/^{137}Cs$ ratio measured from gamma-ray spectrometry) into the isotope ratio equation, it was calculated as 51.5 GWD/MTU. This value shows a good agreement with the chemically obtained result (= 49.3 GWD/MTU) within 4.5 %.

3.4. Pu/U – Isotope Ratio Correlation

^{239}Pu and its isotopes produced by the neutron capture reaction of ^{238}U during a reactor operation accumulate in a fuel and partially contribute to the nuclear fission at the same time. Also ^{134}Cs is produced by the neutron capture reaction of ^{133}Cs. Because both Pu and ^{134}Cs are produced by a neutron capture reaction like this, there is a correlation between Pu and ^{134}Cs for the production quantity. ^{134}Cs/^{137}Cs and Pu/U ratios were calculated by using the results based on the isotopic contents of fission products and actinides for Pu/U ratio determination. These ratios were plotted as shown in Fig.2.
And a correlation equation between the ^{134}Cs/^{137}Cs and Pu/U ratio was derived by using a curve fitting for the data points, it is as follow:

$$IR = -0.03348 + 0.26646(Pu/U) + 0.00279(Pu/U)^2 \quad (2)$$

where IR is the ^{134}Cs/^{137}Cs ratio, and Pu/U is the Pu/U ratio, respectively.

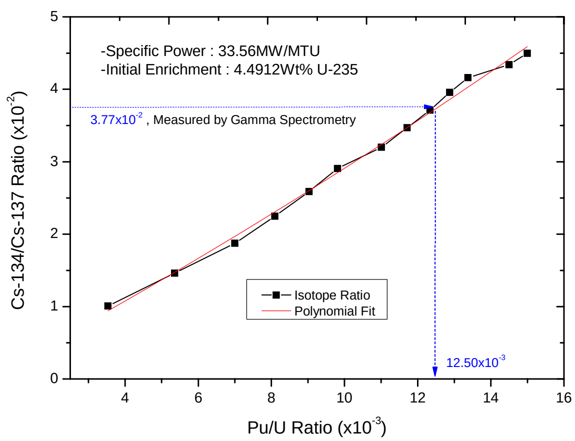

Fig.2. Correlation Curve for the ^{134}Cs/^{137}Cs and Pu/U Ratio.

3.5. Pu/U Ratio Determination

The Pu/U ratio of the J502-R132-2 sample was determined by using the ^{134}Cs/^{137}Cs ratio measured from the gamma-ray spectrometry and the correlation equation (2) derived from the OriginPro-8 graphic software and the ORIGEN-ARP code calculation. As a result of inserting 0.0377 (^{134}Cs/^{137}Cs ratio measured from gamma-ray spectrometry) into equation (2), it was calculated to be 12.50×10^{-3}. This value shows good agreement with the chemically determined one within 0.8% difference as shown in Table 1.

Table 1. Pu/U Ratios determined by Gamma-ray spectrometry and Chemical Analysis

Sample	Axial Position	Isotope Ratio (^{134}Cs/^{137}Cs)	Pu/U Ratio		Diff.
			γ Spectr.	Chemical	
J502-R13-2	2889mm from bot.	0.0377	12.50x10^{-3}	12.6x10^{-3}	0.8%

4. Conclusions

The isotopic ratio of ^{134}Cs/^{137}Cs in a spent PWR fuel sample, J502-R13-2, was obtained with the newly developed gamma/neutron combined measuring system. The burnup and Pu/U ratio of the sample were determined by using the measured isotope ratio and the burnup-isotope ratio correlation equations calculated from the ORIGEN-ARP code. And the results were compared and evaluated by using the chemically determined burnup and Pu/U ratio. As a result of the comparative evaluation, the nondestructively determined burnup and Pu/U ratio values showed a good agreement with the chemically obtained results to within a 4.5 % and 0.8 % difference, respectively.

References

1. I.C. Gauld et al., “ORIGEN-ARP: Automatic Rapid Processing for Spent Fual Depletion, Decay, and Source Term Analysis”, ORNL/NUREG/CSD-2/V1/R7, Volume 1, Revision 7, (2004).