DETERMINATION OF BURNUP AND PU/U RATIO OF PWR SPENT FUELS BY GAMMA-RAY SPECTROMETRY

KWANG-JUNE PARK*, JUNE-SIK JU, JUNG-SUK KIM, HEE-SUNG SHIN, YONG-BUM CHUN and HO-DONG KIM
Korea Atomic Energy Research Institute
P. O. Box 105, Yuseong, Daejeon, 305-600, Korea
*Corresponding author. E-mail : kjpark@kaeri.re.kr

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The isotope ratio of $^{134}Cs/^{137}Cs$ in a spent PWR fuel sample was obtained with a newly developed gamma/neutron combined measuring system at KAERI. Burnup and Pu/U ratio of the spent fuel sample were determined by using the measured isotope ratio and the burnup-isotope ratio correlation equations calculated from the ORIGEN-ARP computer code. The results were compared and evaluated with 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.

KEYWORDS : Burnup, Pu/U Ratio, Isotope Ratio, Gamma-ray Spectrometry, Spent Fuel Sample

1. INTRODUCTION

Nuclear material accounting in spent fuel handling facilities is indispensable to the safeguards and process controls, and various measuring methods are used for it. Several non-destructive assay methods, such as gamma-ray spectrometry, calorimetry, and K-Edge/K-XRF, can be employed in the case of wet processing products without fission products [1]. Especially in the gamma-ray spectrometry, MGA (Multi-Group Analysis) and FRAM (Fixed energy, Response function Analysis with Multiple efficiencies) codes are used in chemical reprocessing plants [2,3]. But if the gamma emitting fission products are included in the processing materials, the above-mentioned methods are limited for a nuclear material accounting. In this case, a burnup referred to as fissile material consumption should be determined by using burnup indicators such as ^{95}Zr, ^{137}Cs, ^{134}Cs, ^{154}Eu, ^{106}Ru, and ^{144}Ce in the spent fuels. Burnup is one of the major factors to identify the safety and economy of nuclear fuel management processes such as storage, transportation, reuse, and disposal of spent nuclear fuels [4-8]. The Pu/U ratio is also a major factor to account for the nuclear materials in the spent fuels. Special nuclear materials such as a separated ^{235}U and ^{239}Pu can be easily counted by various NDA (Nondestructive Assay) methods in the wet reprocessing facility. But we have to determine the quantity of those materials by using the ratio of fission products in the spent fuels because they have not been purely separated in Korea. In this work, a gamma/neutron combined measuring system was newly constructed for a small spent fuel sample application before measuring pyro-processing products. 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. The accuracy of the burnup and Pu/U ratio results obtained from the gamma-ray spectrometry were evaluated by using the chemical analysis results.

2. EXPERIMENTS

2.1 Gamma Detection System Construction

A gamma detection system consists of a HPGe (High Purity Germanium) detector, collimation assembly, and electronic equipment for a spent fuel measurement. A gamma-ray collimator placed between the HPGe detector and a spent fuel sample plays an important role in the shielding and passage of gamma radiation. The detector has to detect all of the incident gamma rays from a disc-typed spent fuel sample (SFS) for a correct data acquisition because of a different isotopic distribution according to the sectional position of the sample. Thus a double cone-shaped collimator as shown in Fig. 1 was devised for the extension of a solid angle and radiation shield. In general, rectangular type collimators with a small slit are mostly used for gamma scanning of disc-typed fuel samples. It takes a lot of time to scan and measure each position of the sample. If we increase the rectangular slit area in order

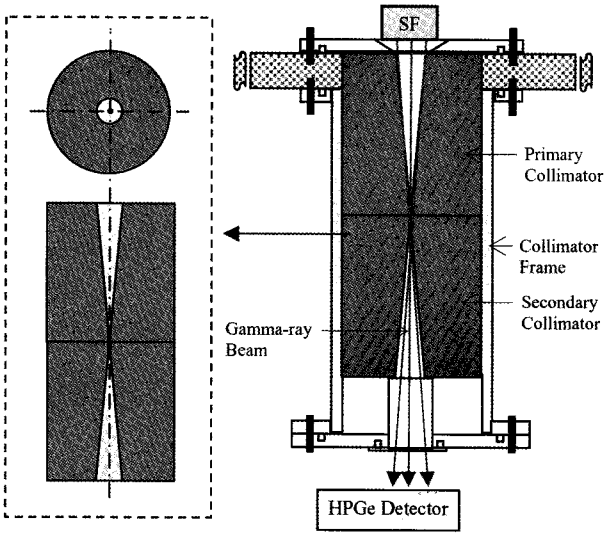

Fig. 1. Schematic Drawing of the Double Cone-shaped Collimator and its Assembly

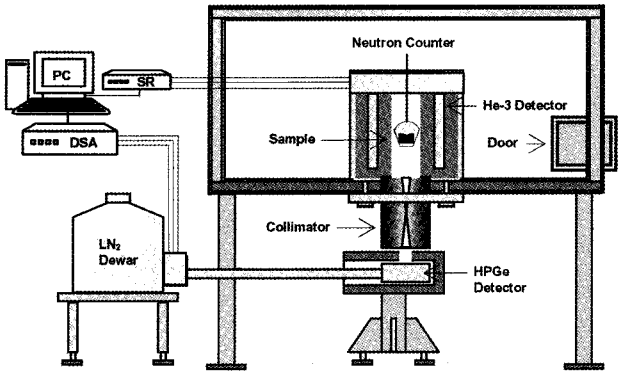

Fig. 2. Schematic Drawing of the Shielded Glove-box for Gamma and Neutron Counter

to cover the total area of the sample, the measuring time can be reduced. But the increases of the gamma count rate are too high due to the increase of the number of gammas emitting from the sample and peaks are crushed. So we have one side of the collimator slit subtended to the sample enlarged more than the other side. It is possible to overcome the demerits of the rectangular collimator because the cone-shaped collimator has a large area slit in the direction of the sample and the small area slit toward the gamma detector. The radiation shielding effect is twice as high when the double cone-shaped collimator is used.

2.2 Shielded Glove-Box

A shielded glove-box was employed for measuring the small spent fuel samples, and a gamma detection system and a neutron counter were installed inside and outside of it, respectively. A transferring cask adapter to be connected to the sample inlet/outlet door was prepared to transfer the spent fuel samples. Fig. 2 shows a schematic drawing of the shielded glove-box installed with the gamma detection system and the neutron counter. A spent fuel sample is positioned in the center of the neutron counter, and gamma photons and neutrons are coincidently detected by the HPGe detector and neutron counter.

2.3 Experimental Procedure

Burnup evaluation tests of spent nuclear fuel assembly/rods and samples were carried out according to the relevant procedures, such as a gamma-ray spectrometry, a computer code calculation, and a chemical analysis as shown in Fig. 3.

First, a gross gamma scanning was carried out along the full length of the fuel. Second, a gamma-ray spectrum was obtained from a specific position, and a specimen for

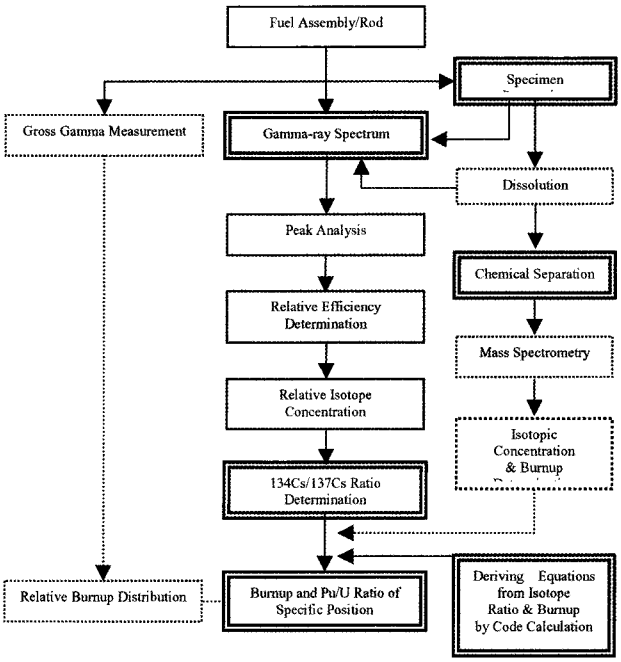

Fig. 3. Burnup and Pu/U Ratio Determination Procedures

the chemical analysis was taken at the next position to the gamma measuring point.

2.3.1 Fuel Rod Gamma Scanning

The total gamma counting rate at each position of the fuel rod is proportional to the burnup. A relative burnup profile of the J502-R13 spent fuel rod was obtained at PIEF (post-irradiation examination facility) as shown in Fig. 4.

2.3.2 Gamma-ray Spectrum Acquisition

Gamma-ray measurement of the J502-R13-2 sample was carried out with a newly developed gamma/neutron

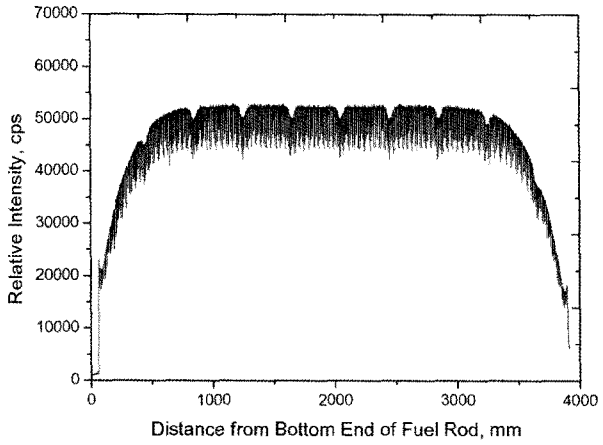

Fig. 4. Relative Burnup Profile of the J502-R13 Fuel Rod

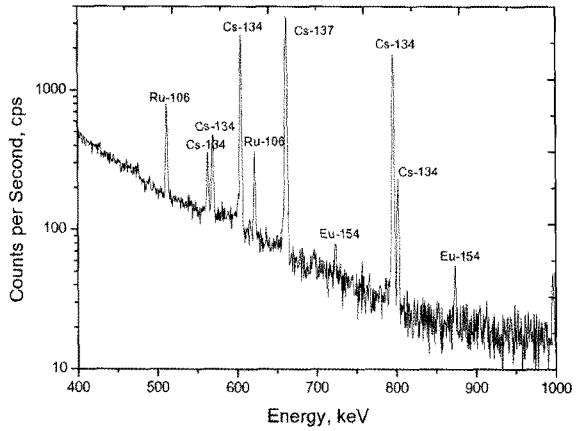

Fig. 5. Gamma-ray Spectrum of the J502-R13-2 Sample

combined system which incorporates GENIE-2000 software for a gamma-ray analysis. The counting time and dead time were 9,000 seconds and 0.94%, respectively. Figure 5 and Table 1 show the gamma-ray spectrum and raw data obtained from the sample.

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.

$$Burnup \cong \frac{N(^{235}U)\sigma_{f235}\phi T}{N(Total\ U)} = E\sigma_{f235}\phi T \propto E\phi_{th}T \qquad (1)$$

Table 1. Gamma Measurement Data of the J502-R13-2 Sample

Isotopes	Energy, keV	Net Peak Area	Net Area Uncert.
^{134}Cs	563.38	2.07×10^{4}	226
	569.40	3.84×10^{4}	266
	604.78	2.80×10^{5}	556
	795.72	3.87×10^{5}	634
	801.78	4.05×10^{4}	230
^{137}Cs	661.63	5.57×10^{5}	853
^{154}Eu	723.19	7.86×10^{3}	269
	873.07	7.48×10^{3}	88
	1004.43	1.09×10^{4}	121
	1274.07	2.49×10^{4}	193
^{106}Ru	511.99	2.94×10^{4}	650
	622.06	2.01×10^{4}	197
^{144}Ce	2185.19	2.52×10^{3}	51

Where, σ_{f235} is the fission cross-section of ^{235}U, ϕ is the neutron flux, T is the neutron-irradiation time, E is the initial enrichment ($=N(^{235}U)/N(TotalU)$), and ϕ_{th} is the thermal neutron flux.

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 as shown in the following equation:

$$\frac{N(^{134}Cs)}{N(^{137}Cs)} \propto \phi_{epi}T \propto Burnup\frac{\phi_{epi}}{E\phi_{th}} \qquad (2)$$

Where, ϕ_{epi} is the epi-thermal neutron flux.

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, and half life. 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.

3.1.1 Relative Detection Efficiency

The absolute or intrinsic full-energy-peak efficiency is frequently not necessary, and only the ratio of the efficiency at different energies is required. In the relative efficiency, exact gamma ray emission rates are not required, only values proportional to the emission rates are required.

When a single multi-energy isotope is used, the branching fractions provide the necessary information. Relative efficiencies can be expressed for an isotope with several gamma energies [9]:

$$\varepsilon_R = \frac{A_i}{Br_i} \tag{3}$$

Where, Br_i is the branching ratio corresponding to the peak area A_i. Usually relative efficiency curves are normalized to 1.00 at a convenient energy range or point.

Taking into consideration the 3.25 years of cooling time of the J502 spent fuel and the half life of ^{134}Cs, five gamma energies of the ^{134}Cs isotope, obtained from the peak analysis in the measured spectrum of the J502-R13-2 sample, were used for the relative efficiency curve determination in this work. Table 2 lists the gamma-ray energies, net peak area, and branching ratios for determining the relative efficiency curve. The gamma-ray branching ratio values were cited from the Atomic Data & Nuclear Data Tables [10].

Table 2. Data for Relative Efficiency Curve Determination

Isotope	Energy, keV	Net peak area	Branching ratio
^{134}Cs	563.38	2.07×10^4	8.40%
	569.40	3.84×10^4	15.00%
	604.78	2.80×10^5	97.54%
	795.72	3.87×10^5	85.13%
	801.78	4.05×10^4	8.80%

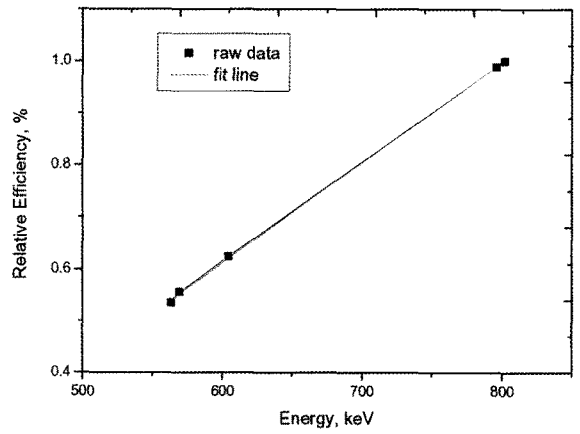

Fig. 6. Relative Efficiency Curve of HPGe Gamma-ray Detection System

The relative efficiency according to the five energies was calculated by putting the data of Table 2 into equation (3), and then the relative efficiency curve was determined by fitting the calculated efficiencies as shown in equation (4) and Fig. 6.

$$\varepsilon_R = -0.54409 + 0.00193E_i \tag{4}$$

Errors at the intercept and the first order's coefficient are 0.01230 and 0.00002, respectively. The adjust R-Square (coefficient of determination) is 0.99964.

3.1.2 Atomic Density

Generally, the activity emitted from fission products is represented as follows:

$$\lambda_i N_i = \frac{A_i}{Br_i \varepsilon(E_i)} \tag{5}$$

Where, $\lambda_i(=\ln 2/T_{1/2})$ is a decay constant of isotope i, $T_{1/2}$ is a half life of isotope i, N_i is the number of atoms of isotope i, A_i is the net peak area of isotope i, Br_i is the gamma-ray energy branching ratio of isotope i, and $\varepsilon(E_i)$ is the detection efficiency of the gamma ray energy E_i. The number of atoms of isotope i, N_i, can be expressed as follows using equation (5):

$$N_i = \frac{A_i}{(\ln 2/T_{1/2})Br_i \varepsilon(E_i)} \tag{6}$$

The isotope ratio between isotopes A and B can be written in the following form:

$$\frac{N_B}{N_A} = \frac{A_B}{A_A}\frac{T_{1/2(B)}}{T_{1/2(A)}}\frac{Br_A}{Br_B}\frac{\varepsilon(E_{i(A)})}{\varepsilon(E_{i(B)})} \tag{7}$$

The isotope ratio of ^{134}Cs/^{137}Cs in this work was determined by using equation (7), the net peak area of Table 1, and the branching ratio and half life of ^{134}Cs and ^{137}Cs in the Atomic Data & Nuclear Data Tables [10]. Also, the relative efficiency in equation (3), ε_R, was applied in equation (7) instead of the absolute full-energy peak efficiency $\varepsilon(E)$ because of the isotope ratio calculation. As a result of the calculation, the value of the ^{134}Cs/^{137}Cs ratio was calculated to be 0.037.

3.2 Burnup-Isotope Ratio Correlation

It is not easy to directly determine a spent fuel burnup

Table 3. Input Data for Isotope Ratio Calculation and $^{134}Cs/^{137}Cs$ Ratio

Isotope	E (keV)	Peak area	Half-life (year)	Br (%)	ε_R	$\frac{N_{134}}{N_{137}}$
^{134}Cs	604.78	2.80×10^5	2.06	97.54	0.62	0.037
^{137}Cs	661.63	5.57×10^5	30.00	89.93	0.73	

Table 4. Major Isotope Contents According to the Burnup Variation

Burnup, MWD/MTU	^{134}Cs, g	^{137}Cs, g	^{154}Eu, g	^{106}Ru, g
10,000	3.6	356.0	-	3.7
15,000	7.8	530.9	-	5.9
20,000	13.2	703.5	-	8.1
25,000	19.7	876.2	-	10.3
30,000	27.1	1046.0	13.4	12.4
35,000	35.3	1213.0	17.9	14.5
40,000	44.1	1378.0	22.6	16.5
45,000	53.4	1541.0	27.4	18.5
50,000	63.2	1703.0	32.2	20.4
55,000	73.7	1862.0	36.9	22.1
60,000	84.0	2019.0	41.5	23.8
65,000	94.4	2174.0	45.7	25.4
70,000	104.6	2326.0	49.5	26.9

by a gamma-ray spectrometry, so a correlation between burnup and isotope ratio that is able to be achieved by a code calculation is needed.

3.2.1 ORIGEN-ARP Calculation

The number of fissile materials and fission products during a reactor operation varies with the initial enrichment, fuel type, specific power, burnup, etc. The ORIGEN-ARP (Automatic Rapid Processing for spent fuel depletion, decay, and source term analysis) code [11] was used to establish the ratio of isotopes according to the burnup variation from 10,000 MWD/MTU to 70,000 MWD/MTU at the interval of 5,000 MWD/MTU. The input data for the J502 fuel were 17×17 UO_2 fuel type, 18 group SCALE in gamma energy, 27 group ENDF4 in neutron energy, an initial enrichment of 4.4912 wt% ^{235}U, a specific power of 33.56 MW/MTU, and an uranium weight of 1,000,000 g. Table 4 shows the results of the code calculation for some isotopes.

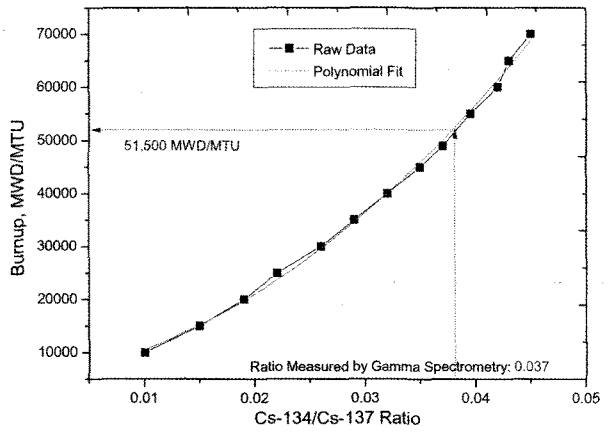

Fig. 7. Correlation Curve for the Burnup-$^{134}Cs/^{137}Cs$ Ratio of J502 Spent Fuel

3.2.2 Correlation Equation

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 on the graph as shown in Fig. 7. The correlation equation is as follows

$$Burnup = 5169.82 + 295118.99(IR) + 2.48\times10^7(IR)^2 \quad (8)$$

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

Errors at the intercept, and the first and second order's coefficients are 1961.58, 150794.27, and 2.62557×10^6, respectively. The adjusted R-Square (determination coefficient) is 0.9974.

3.2.3 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 (8) derived from the results of the ORIGEN-ARP code calculation and the OriginPro-8 graphic software. As a result of inserting 0.037 ($^{134}Cs/^{137}Cs$ ratio measured from gamma-ray spectrometry) into equation (7), it was calculated as 51,500

Table 5. Burnup Values Determined by the Gamma-ray Spectrometry and the Chemical Analysis

Sample	Axial Position	Isotope Ratio ($^{134}Cs/^{137}Cs$)	Burnup, MWD/MTU		Diff.
			γ Spectr.	Chemical	
J502-R13-2	2889mm from bottom	0.037	51,500	49,300	4.5%

Table 6. Plutonium Contents in the J502-R13-2 Sample

Sample	Burnup, MWD/MTU	Pu content, mg-Pu/g-sample					
		Total	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
J502-R13-2	49,300±1,600	10.05 (0.29)	0.33 (0.01)	5.14 (0.15)	2.51 (0.07)	1.31 (0.04)	0.76 (0.02)

*The figure of parenthesis refers to the error.

Table 7. Uranium Contents in the J502-R13-2 Sample

Sample	Burnup, MWD/MTU	U content, mg-U/g-sample				
		Total	^{234}U	^{235}U	^{236}U	^{238}U
J502-R13-2	49,300±1,600	799 (23.00)	0.20 (0.01)	7.72 (0.22)	5.32 (0.16)	786.00 (22.00)

*The figure of parenthesis refers to the error.

MWD/MTU. This value shows a good agreement with the chemically obtained result (= 49,300 MWD/MTU) within 4.5%.

3.2.3 Chemical Burnup

The burnup in fissions per initial metal atom (FIMA) is chemically determined by the Nd-148 method (ASTM-E321) [12] and expressed as follows:

$$Burnup = \frac{^{148}Nd/U/Y_{148}}{1 + Pu/U + Np/U + Am/U + Cm/U + ^{148}Nd/U/Y_{148}} \quad (9)$$

Where, $^{148}Nd/U$, Np/U, Pu/U, Am/U, and Cm/U are the measured atom ratios, Y_{148} is the effective fission yield of ^{148}Nd, which is obtained by using fission fractions of ^{235}U, ^{238}U, ^{239}Pu, and ^{241}Pu, calculated by manual work, and their yields for thermal neutron (ENDF-349, 1994) [13]. Burnup in MWD/MTU is obtained by multiplying the value in % FIMA by 9,600.

Table 6 and Table 7 show the results of the burnup and the Pu and U contents determined from the chemical analysis of the spent fuel sample, J502-R13-2.

3.3 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 the 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.

3.3.1 ORIGEN-ARP Calculation

In this context, the Pu/U ratio was calculated by the ORIGEN-ARP code. Generally, the Pu/U ratio varies with a specific power, but it is not much affected by the initial enrichment. In this calculation, the specific power and initial enrichment were fixed as 33.56 MW/MTU and 4.4912 wt% ^{235}U, respectively. Table 8 shows the contents of some fission products and actinides for the Pu/U determination as a result of the code calculation.

3.3.2 Correlation Equation

$^{134}Cs/^{137}Cs$ and Pu/U ratios were calculated using the results based on Table 8. These ratios were plotted on the graph as shown in Fig. 8.

A correlation equation between the $^{134}Cs/^{137}Cs$ and Pu/U ratios was derived using a curve fitting for the data points. It is as follow:s

$$IR = -0.03348 + 0.26646(Pu/U) + 0.00279(Pu/U)^2 \quad (10)$$

Where, *IR* is the $^{134}Cs/^{137}Cs$ ratio, and is the *Pu/U* ratio, respectively. Errors at the intercept, and the first and second order's coefficients are 0.16864, 0.03784, and 0.00196, respectively. The adjusted R-Square (coefficient of determination) is 0.995.

Table 8. Isotopic Contents of Fission Products and Actinides for Pu/U Ratio Determination

Burnup, MWD/MTU	^{134}Cs, g	^{137}Cs, g	Pu, g	U, g
10,000	4	356	3.477×10^{3}	9.849×10^{5}
15,000	8	531	5.237×10^{3}	9.781×10^{5}
20,000	13	704	6.799×10^{3}	9.715×10^{5}
25,000	20	876	7.811×10^{3}	9.650×10^{5}
30,000	27	1046	8.656×10^{3}	9.588×10^{5}
35,000	35	1213	9.343×10^{3}	9.526×10^{5}
40,000	44	1378	1.042×10^{4}	9.465×10^{5}
45,000	53	1541	1.102×10^{4}	9.406×10^{5}
50,000	63	1703	1.153×10^{4}	9.347×10^{5}
55,000	74	1862	1.196×10^{4}	9.288×10^{5}
60,000	84	2019	1.235×10^{4}	9.231×10^{5}
65,000	94	2174	1.330×10^{4}	9.174×10^{5}
70,000	105	2326	1.367×10^{4}	9.118×10^{5}

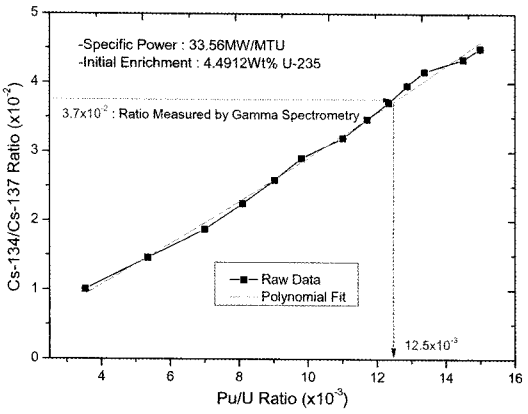

Fig. 8. Correlation Curve for the $^{134}Cs/^{137}Cs$ and Pu/U Ratios

3.3.3 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 (10) derived from the OriginPro-8 graphic software and the ORIGEN-ARP code calculation. As a result of putting 0.037 ($^{134}Cs/^{137}Cs$ ratio measured from gamma-ray spectrometry) into equation (10), 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 9.

4. CONCLUSIONS

A gamma/neutron combined counter was designed for a small spent fuel sample application at KAERI for the purpose of a nuclear material accounting study. In this work, only a gamma-ray spectrometer was used for the measurement of the spent fuel sample.

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 using the measured isotope ratio and the burnup-isotope ratio correlation equations calculated from the ORIGEN-ARP code. The results were compared and evaluated 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.

Although these results were determined from a specific position of the spent fuel rod, the average burnup and Pu/U ratio of the entire fuel rod or assembly can be determined by using the gross gamma scanning data obtained along the full length of it. This nondestructive gamma method will be able to be applied to the nuclear material accounting of spent nuclear fuels along with the neutron method instead of the time-consuming and expensive chemical analysis with enough experiences for the various fuel types, enrichment, specific power, and burnup in the future.

ACKNOWLEDGEMENT

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Table 9. Pu/U Ratios Determined by Gamma-ray Spectrometry and Chemical Analysis

Sample	Axial Position	Isotope Ratio (134Cs/137Cs)	Pu/U Ratio		Diff.
			γ Spectrometry	Chemical	
J502-R13-2	2889mm from bottom.	0.037	12.5×10^{-3}	12.6×10^{-3}	0.8%

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