DETERMINATION OF BURNUP, COOLING TIME AND INITIAL ENRICHMENT OF PWR SPENT FUEL BY USE OF GAMMA-RAY ACTIVITY RATIOS

D.K. MIN, H.J. PARK, K.J. PARK, S.G. RO, H.S. PARK
Korea Atomic Energy Research Institute,
Yusong , Taejon,
Republic of Korea

1. INTRODUCTION

The burnup, cooling time and initial enrichment of spent nuclear fuel are basic parameters to characterize the radionuclide inventory, decay heat output and fissile material of the spent fuel. It is very important to determine these parameters with a high level of confidence, because of its impact on safety and costs during spent fuel management operations, such as storage, transportation, reprocessing and disposal. For the non-destructive determination of these spent fuel parameters, radiometric methods have been developed in the USA, Europe and Japan. These methods are based on a combination of the neutron emission and gamma measurement techniques.

The Korea Atomic Energy Research Institute (KAERI) has been developing the algorithms for the sequential determination of cooling time, initial enrichment and burnup of the pressurized water reactor (PWR) spent fuel assembly by only use of the gamma ratios measurements, i.e., $^{134}Cs/^{137}Cs$, $^{154}Eu/^{137}Cs$ and $^{106}Ru\ ^{137}Cs/(^{134}Cs)^2$. This method has advantages over combination techniques of neutron and gamma measurement, because of its simplicity and insensitivity to the measurement geometry.

For verifying the algorithms developed, an experiment for determination of cooling time, initial enrichment and burnup of the two PWR spent fuel rods have been conducted by use of the high-resolution gamma detector (HPGe) system only. This paper describes the method used and interim results of the experiment.

2. METHOD

Fig. 1 shows the flow chart for the burnup measurement algorithm used in this work. As shown in Fig. 1, the algorithm can be divided into three parts, activity ratio calculation including the regression analysis of the activity ratios, the activity ratio measurements by gamma spectroscopy and the determination of the spent fuel parameters such as the cooling time, initial enrichment and the burnup.

2.1. ORIGEN-S calculations

Activity ratios of $^{134}Cs/^{137}Cs$, $^{154}Eu/^{137}Cs$, and $^{106}Ru^{137}Cs/(^{134}Cs)^2$ in PWR spent fuel were calculated using the ORIGEN-S code with various spent fuel parameter input data; (1) enrichment (2.5~5.0 wt%), (2) cooling time (0~20 years), (3) burnup (10~60 GW·d/tU), and (4) fuel type (14x14, 16x16, 17x17). With the regression analyses of the calculation results, the activity ratios were correlated as a function of burnup (*bu*), cooling time after irradiation (*t*), and initial enrichment (*en*).

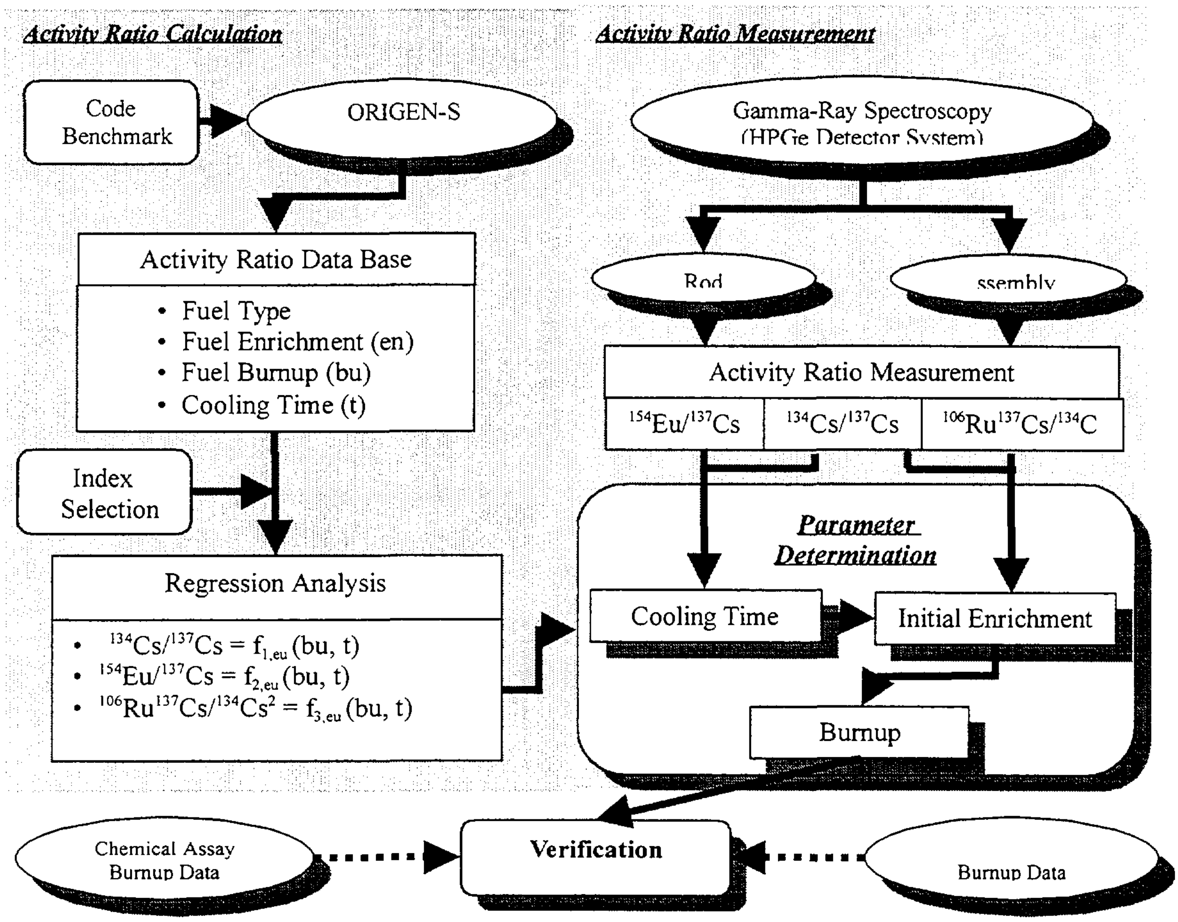

FIG. 1. Flow chart for the burnup measurement algorithm

$$^{134}Cs/^{137}Cs = \{P_1(en) + P_2(en)\cdot bu + P_3(en)\cdot bu^2\}e^{-(\lambda_{34}-\lambda_{37})t} \quad (1)$$

$$^{154}Eu/^{137}Cs = \{Q_1(en) + Q_2(en)\cdot bu + Q_3(en)\cdot bu^2\}e^{-(\lambda_{54}-\lambda_{37})t} \quad (2)$$

$$\ln[^{106}Ru\ ^{137}Cs/(^{134}Cs)^2] = R_1(en) + R_2(en)\cdot \ln(bu) - (\lambda_{06} + \lambda_{37} - 2\lambda_{34})t \quad (3)$$

where *Pi(en)*, *Qi(en)* and *Ri(en)* are the regression coefficient at initial enrichment *en* and the activity ratio *i*.

2.2. Cooling time determination

At first, the burnup of the spent fuel was calculated with various cooling times and initial enrichments, putting the measured isotope ratios of $^{134}Cs/^{137}Cs$ and $^{154}Eu/^{137}Cs$ into Eqs. (1) and (2). The difference of the burnup calculated by Eqs. (1) and (2) should be minimal at the true values of the cooling time and the initial ^{235}U enrichment. For the computational analysis of the burnup differences, an estimator F was introduced which is defined as:

$$F \equiv \frac{1}{N}\sum_{i=1}^{N}\frac{|\ B_i(Cs) - B_i(Eu)\ |}{\overline{B_i}} \quad (4)$$

where i = *1* to N is the index for the gamma ray measurement positions and $B_i(Cs)$ and $B_i(Eu)$ are the measured burnup using the activity ratios of $^{134}Cs/^{137}Cs$ and $^{154}Eu/^{137}Cs$, respectively. Therefore, F is an average fractional difference between the burnup calculated using $^{134}Cs/^{137}Cs$ and

$^{154}Eu/^{137}Cs$. The characteristics of *F* are insensitive on initial enrichment variations and exponentially dependent on the cooling time. Using these characteristics, the cooling time of spent fuel can be very sensitively determined as the value which minimizes *F* with the given initial enrichment ranges [1].

2.3. Initial enrichment determination

With known cooling time of the spent fuel, the initial enrichment can be determined in a similar way as in the cooling time determination step, using measured gamma-ray activity ratios of $^{134}Cs/^{137}Cs$ and $^{106}Ru^{137}Cs/(^{134}Cs)^2$.

For the determination of the initial enrichment, another estimator *F'* is introduced, defined as:

$$F' \equiv \frac{1}{N}\sum_{i=1}^{N}\frac{|\, B_i(Cs) - B_i(Ru)\,|}{\overline{B_i}} \tag{5}$$

where $B_i(Ru)$ is the measured burnup using the activity ratio of $^{106}Ru^{137}Cs/(^{134}Cs)^2$.

Using the cooling time (previously determined) and the measured gamma-ray activity ratios of $^{134}Cs/^{137}Cs$ and $^{106}Ru^{137}Cs/(^{134}Cs)^2$, one can plot the *F'* value as a function of the initial enrichment with Eqs. (1), (3) and (5). The initial enrichment can be determined as the time value of the minimum *F'* value.

2.4. Burnup determination

If the cooling time and the initial ^{235}U enrichment are determined, one can easily determine the burnup at each measurement position using Eqs. (1)-(3) [2].

3. EXPERIMENT

The gamma-ray spectrometric experiments were carried out on the two PWR spent fuel rods using a high purity Ge(HPGe) detector system installed in the KAERI hot cells. The first rod (Rod A) irradiated in Kori Unit-1 (14x14 fuel type) has a discharge burnup of 39 GW·d/tU (operator declared rod average burnup), a initial enrichment of 3.2 wt% and a cooling time of 8.45 years as of measurement time. While the other rod (Rod B) irradiated in Kori Unit-2 (16x16 fuel type) has a discharge burnup of 35 GW·d/tU, a initial enrichment of 3.5 wt% and a cooling time of 5.3 years as of measurement time (see Table I).

TABLE I. SPECIFICATION OF EXAMINED SPENT FUEL

Spent Fuel	Discharged Date	Initial Enrichment	Operator Declared Burnup
	(Cooling time)	(wt%)	(GWd/tU)
Rod A (14x14 PWR)	January 1989 (8.5 years)*	3.2	39
Rod B (16x16 PWR)	May 1992 (5.3 years)*	3.5	36

* As of measurement time.

4. RESULTS

4.1. Cooling time

Fig. 2 shows the cooling time as a function of the initial enrichment. This figure indicates a very weak correlation between initial enrichment and the cooling time. The average cooling time of Rod A and B over initial enrichments from 2.5 to 5.0 wt% were calculated to be 8.76 years and 5.22 years, respectively. These values are in good agreement with the operator declared cooling times within 2.5 %.

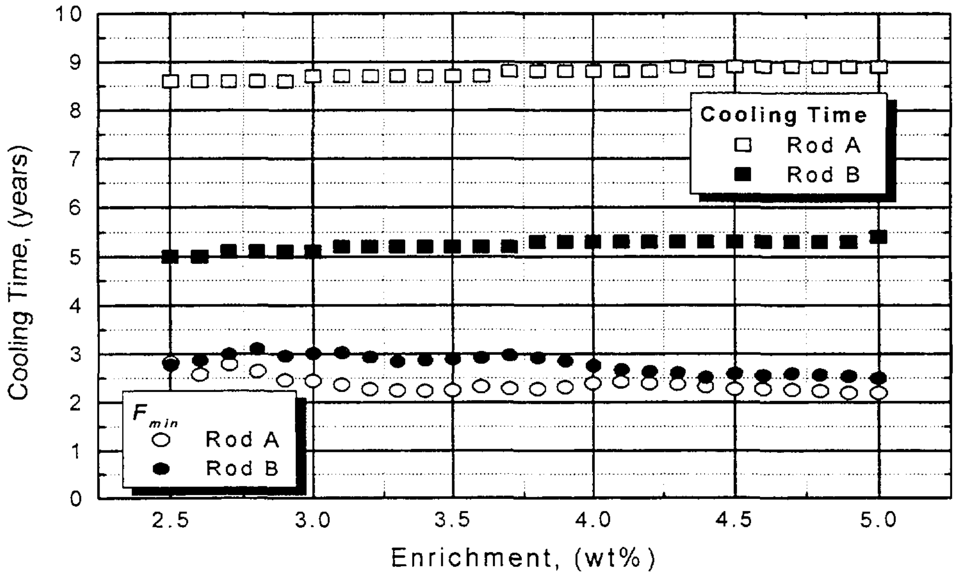

FIG. 2. Cooling time as a function of initial enrichment

4.2. Initial enrichment

Fig. 3 shows an estimator *F'* of Rod A and B as a function of initial enrichments with the previously determined cooling time of 5.22 years. It indicates, that *F'* of Rod B is minimum at 3.6 wt% initial enrichment, i.e., the initial enrichment of Rod B was determined to be 3.6 wt%, which is in good agreement with the operator declared initial enrichment of 3.5 wt% within 2.9 %. However, the determination of the initial enrichment of Rod A failed. The main reason is likely caused by the uncertainty from the ^{106}Ru inventory from the ORIGEN-S calculation.

4.3. Burnup

Putting the cooling time and initial enrichment value previously determined into Eq. (2), the averaged burnup of Rod A and B were easily determined to be 41.3 and 34.6 GW·d/tU, respectively, which differ from the operator declared burnup of 39 and 35 GW·d/tU by 5.9 and 1.1% (see Fig. 4).

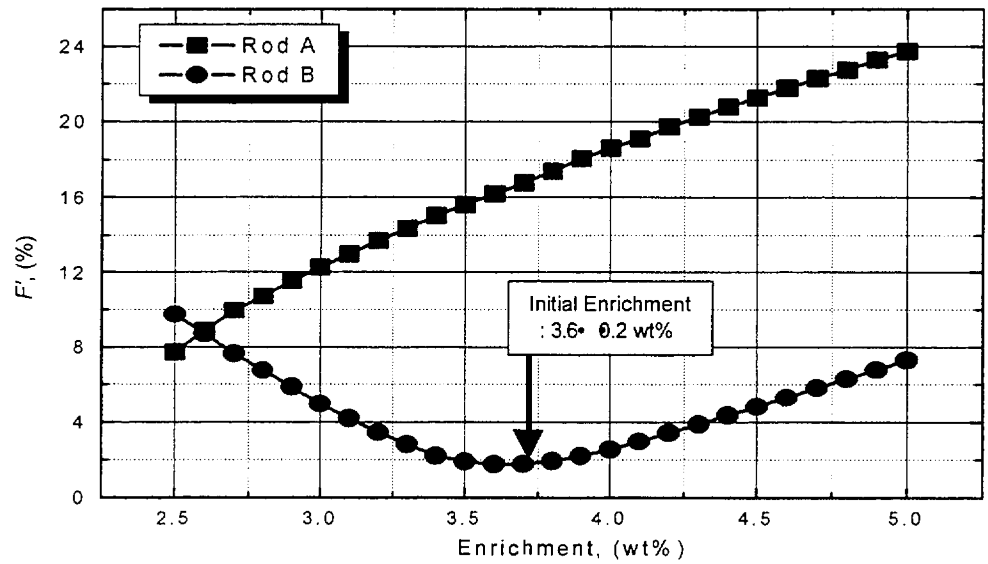

FIG. 3. Initial enrichment determination using estimator F's of Rod A and B.

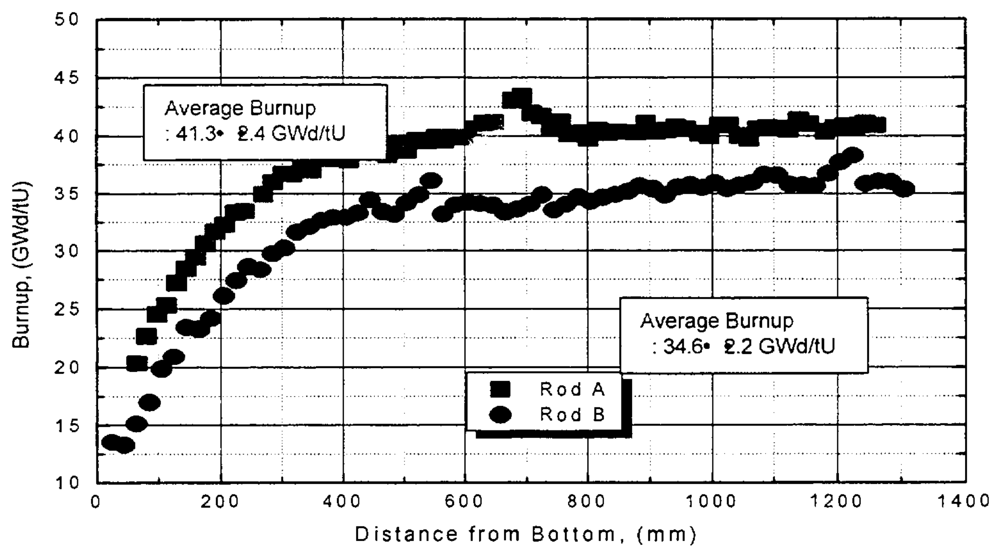

FIG. 4. Burnup distribution of Rod A and B

5. SUMMARY

Experimental results for PWR rod A and B are summarized in Table II. It indicates that the method (or algorithms) has a good reproducibility, i.e., within 2.5 % for the cooling time and 6 % for the burnup. However, the initial enrichment of Rod A was not determined, probably due to the uncertainty of the ^{106}Ru inventory from ORIGEN-S calculation.

It is preliminary demonstrated that this technique is a very useful tool in burnup determination without information on the cooling time and initial enrichment of the spent fuel. This technique can be applied to spent fuel characterization, burnup credit and safeguards of the spent fuel management facility.

For the evaluation of the fuel assembly burnup, further examinations of the PWR spent fuel assembly, improvement of the algorithms including the ^{106}Ru benchmarking and the axial/radial averaging routines are required.

TABLE II. RESULTS OF THE COOLING TIME, THE INITIAL ENRICHMENT AND THE BURNUP OF PWR SPENT FUEL RODS A AND B.

Rod ID	Cooling Time (years)	Initial Enrichment (wt%)	Burnup (GW·d/tU)
Rod A			
- Measured	8.76 (2.4%)*	-	41.3 (5.9 %)
- Operator declared	8.45	3.2	39
Rod B		5	
- Measured	5.22 (-1.9 %)	3.6 (2.9 %)	34.6 (-3.9 %)
- Operator declared	5.3	3.5	35

*: Numbers in parenthesis stands for the differences in % between the measured and the operator declared values.

ACKNOWLEDGEMENT

This work has been carried out under the R&D Programme of Ministry of Science and Technology (MOST), Korea.

REFERENCES

[1] PARK, H. J., MIN, D.K., et al.," Cooling time determination of PWR spent fuel using gamma-ray activity ratios of $^{134}Cs/^{137}Cs$ and $^{154}Eu/^{137}Cs$," *Proc. of Kor. Nuc. Soc. Spring Meeting* (Seoul, Korea, 1998), Vol. II, (1998) 545,550.

[2] PARK, H. J., MIN, D.K., et al.," Burnup determination of PWR spent fuel using gamma-ray activity ratio", *Proc. of Kor. Nuc. Soc. Spring Meeting* (Seoul, Korea, 1998), Vol. II, (1998) 509,514.