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Burnup Estimation of Nuclear Fuels with Gamma Spectrometry

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1. Introduction

Nondestructive tests have been performed on spent nuclear fuel to verify burnup and cooling time for safeguard purposes. In gamma scanning test, Cs-137 is used as a monitor nuclide for the burnup[1]. The additional fission products studied are Cs-134 and Eu-154, these isotopes dominate the gamma-ray spectrum of a spent fuel with a cooling time of 10-20 years[2].

Without a geometry correction, the activity ratios of the nuclides, which are fission products, have been introduced. Haddad calculated the cooling time of the fuel with the activity ratio of Nb-95/Zr-95[3]. Iqbal and et al. performed a gamma scanning to obtain the activity ratios of Zr-95/Cs-137 and Cs-134/Cs-137[4].

For a burnup calculation, the ORIGEN-ARP code is available to calculate the atomic ratio instead of the activity ratio as a burnup calculation[5,6].

Whenever the fuel capsule irradiation in some test holes of HANARO research reactor, HANAFMS, the reactor core calculation code, has been used for the burnup calculation, but the burnup calculation is needed to be verified.

In this study, we tried to verify the burnup calculated by HANAFMS using the gamma scanning test.

2. Methods and Results

PWR spent fuel from nuclear power plant and two irradiated fuel capsules were prepared for burnup estimation. The former declared burnup was used as preliminary test. The latter was for burnup estimation. Two fuel capsules(02F-11K, 03F-05K) containing each three fuel rigs were irradiated in the HANARO research reactor. then a gamma scanning was carried out.

PWR spent fuel rod with 40,000 MWd/t-U and cooling time of 17 years was cut several parts and 10 pellets were taken out and contained in small CANDU fuel tube. This rod containing 10 pellets was moved to the gamma spectrometry system to carry out gamma scanning test.

On the other hand, three fuel rigs of each capsule were made for an irradiation and temperature test[7]. Each rig has 5 UO_2 pellets. 02F-11K was irradiated at the OR5 hole in the HANARO research reactor in March, 2003 and 03F-05K in April, 2004. These capsules had a different linear power history for 54 days and 60 days, then cooled down for 20 months and 12 months, respectively.

After being cooled down, the capsules were dismantled to withdraw the fuel rigs. The gamma scannings for all the fuel were performed with the geometry as shown in Fig. 1. To measure the center temperature of fuel rigs in irradiated capsules, 3 pellets from the top were drilled for a thermocouple. The slit dimension of the collimator is 40 mm(W) x 2 mm(H) x 250 mm(D) and it is made of tungsten. The detecting distance was 160 cm due to a 100 cm wall thickness. The three detecting positions of each fuel rig were selected as shown in figure, which were the first, third and fifth pellets from the top of the rig while one point of PWR spent fuel was selected.

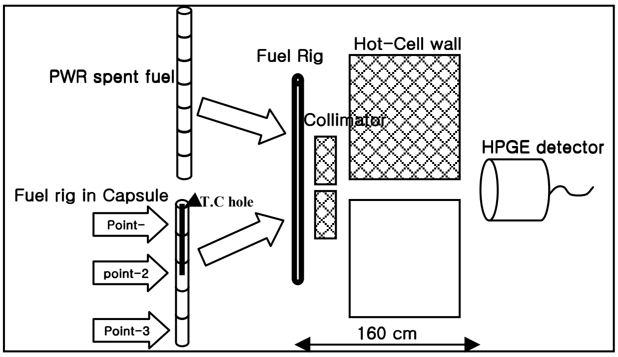

Figure 1. Schematic drawing of gamma scanning for PWR spent fuel and fuel rigs in two irradiated capsules.

After setting up the detecting points, the gamma scannings were performed for 3 hours at a each point. They were carried out every three times at each point repeatedly to reduce the counting errors of the cesium isotopes.

Atomic ratio of Cs-134/Cs-137, instead of the activity ratio, was applied in this study while the gamma peak of Eu-154 in fuel rigs was too small due to a short irradiation time. To calculate the atomic ratio of Cs-134/Cs-137, the basic equation is as follows;

$$C(E_i) = \lambda N P(E_i)\varepsilon(E_i) \qquad (1)$$

Where, C is the gamma counts(cps) at each energy, λ is the decay constant(s^{-1}), N is the atomic amount, P is the decay branch ratio, and ε is the total detector efficiency Here, N. is the radioactivity. Most papers have shown the activity ratio(A_{134}/A_{137}), but we show the function of (N_{134}/N_{137}) via function of Nε with a different viewpoint as shown in eq.(2).

$$\frac{N_{134}\varepsilon(E_{134})}{N_{137}\varepsilon(662keV)} = \frac{C_{134}/(\lambda_{134}P_{134})}{C_{137}/(\lambda_{137}P_{137})} \quad (2)$$

From Eq.(2), the numerator in the left term can be changed to $N_{134}\varepsilon$(662keV) by means of an interpolation of the plot of $N_{134}\varepsilon$ (E_{134}) at 662 keV.

ORIGEN-ARP code was used to calculate burnup which are related the atomic ratio obtained experiments. Fig.2 shows atomic ratio with burnup at each capsule following the code calculation.

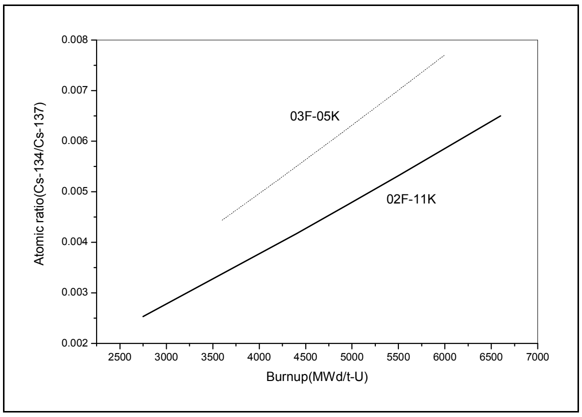

Figure 2. The plot of the atomic ratio with the burnup by ORIGEN-ARP.

The atomic ratio for PWR spent fuel rod was $5.84\text{x}10^{-4}$ and the related burnup from ORIGEN-ARP was 39,500 MWd/t-U. It gave good agreement compared to declared burnup of 40,000 MWd/t-U. In the case, code library was correctly applied as PWR spent fuel. Table.1 shows the atomic ratios and burnups in each capsule.

Table 1. The atomic ratios of Cs-134/Cs-137 and burnups at each rig

Position			Point-1	Point-2	Point-3
02F-11K	Rod #1	R_{exp}	0.0055	0.0051	0.0050
		Bu	5.66	5.3	5.25
	Rod #2	R_{exp}	0.0055	0.0051	0.0050
		Bu	5.66	5.3	5.25
	Rod #3	R_{exp}	0.0044	0.0042	0.0042
		Bu	4.62	4.4	4.4
03F-05K	Rod #1	R_{exp}	0.0060	0.0053	0.0050
		Bu	4.7	4.2	4.0
	Rod #2	R_{exp}	0.0073	0.0065	0.0060
		Bu	5.75	5.15	4.79
	Rod #3	R_{exp}	0.0071	0.0061	0.0059
		Bu	5.58	4.9	4.7

All of rigs show that burnup decreased vertically from top to bottom due to the neutron flux shape. One of three rigs in each capsule had lower burnup in particular. Anyway, the burnups of HANAFMS were 5.9 MWd/t-U for 02F-11K and 5.6 MWd/t-U for 03F-05K. Data from gamma scanning test were lower than those from HANAFMS with an error of 15%.

3. Conclusion

we tried to verify burnup from HANAFMS using a gamma scanning test. Fuel capsules(02F-11K, 03F-05K) containing three 3 UO_2 fuel rigs were irradiated with a different linear power history in the HANARO research reactor for 54 days and 60 days, respectively. Gamma scannings were performed to obtain the Cs-134 and Cs-137 peaks for the atomic ratio of Cs-134/CS-137. The atomic ratio would be better than the activity ratio in the point of a time-consuming for the detector efficiency calibration. Burnup was obtained by ORIGEN-ARP code with the given atomic ratio from the gamma scanning. Preliminary test for PWR spent fuel declared burnup was carried out to compare with data of ORIGEN-ARP before capsule test. It gave good agreement following PWR library applied to code. In the capsules, burnups were 5.1 GWd/MTU for the 02F-11K and 4.8 GWd/MTU for the 03F-05K as mean value. These results were lower than those of HANAFMS by 15% due to the CANDU library applied to ORIGEN-ARP incorrectly. It is necessary to develop the new library for HANARO irradiation hole.

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