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Burnup Evaluation of Spent PWR Fuel by Measuring Gamma-Ray of Fission Product Cs-137

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핵분열 생성핵종 Cs-137 감마선의 측정에 의한 PWR 사용후 핵연료 연소도 평가

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Abstract

Spent PWR fuel rods have been scanned axially and sectionally to measure the relative gamma-ray intensity of Cs-137 and then burnups of the scanned rods determined by measuring Nd-148 which has been chemically separated. From these experimental results, a linear relation(LR) between the gamma-ray intensity of Cs-137 and the burnup in the range of 10~35 GWD/MTU was obtained. In order to validate the LR, the Cs-137 gamma-ray intensity of unknown sample was nondestructively measured and the burnup obtained by the LR was compared with that of the Nd-148 method.

It is revealed that the results from both methods are in good agreement, and thus it seems to be possible to estimate the burnup of spent PWR fuel rod by measuring nondestructively gamma-ray of fission product Cs-137.

요 약

사용후 가압경수로 핵연료봉속에 들어 있는 핵분열 생성핵종의 하나인 Cs-137 감마선의 상대적 세기를 핵연료봉의 길이방향 및 단면방향으로 측정(스캐닝)하고 이곳에서 시료를 취하여 화학분석법인 Nd-148 정량법으로 연소도를 구한 후 Cs-137 검출세기와 연소도간의 관계식을 유도하였는데 이들은 연소도 영역 10~35 GWD/MTU에서 일차 선형적으로 비례하고 있음이 확인되었다. 아울러, 임의의 사용후 가압 경수로 핵연료를 대상으로 한 검증시험에서 검출된 Cs-137 감마선의 세기를 이 관계식에 대입하여 계산한 연소도와 Nd-148 정량법으로 산출한 값을 비교하였다.

그 결과, 양자간에는 잘 일치하고 있음을 보여주었다. 이로서 유도된 관계식을 이용하여 Cs-137 감마선세기의 분포로 부터 연소도를 추정하는 것이 가능하게 되었다.

I. Introduction

Method employing radioactive fission products as burnup monitors of spent nuclear fuel has been in use widely for spent fuel management and safeguards purposes[1]. Burnup can be estimated by analyzing gamma-ray spectra which are obtained from relatively long-lived fission products such as Zr-95, Ru-106, Cs-137 and Ce-144. There are advantages and disadvantages in each burnup monitor[2-4].

Cs-137($T_{1/2}$=30.2 years) is widely used as a burnup monitor because of its easily resolvable gamma-ray peak and its long half-life. This isotope requires no knowledge of neutron spectrum or fuel compositions and almost no knowledge of irradiation history for interpretation because the fission yields for both U and Pu are almost identical as well as its destruction by neutron capture is negligible. The only distinct disadvantage of Cs-137 is its tendency to migrate radially and axially in spent fuel rod under certain conditions.

Ru-106($T_{1/2}$=1.0 year) can be employed as a burnup monitor for Pu or highly enriched fuels because it has a much higher fission yield for Pu than U.

Ce-144($T_{1/2}$=285 days) finds an application for a potential burnup monitor because of its half-life and easily resolvable gamma-ray peaks. For long irradiations, the half-life becomes a distinct disadvantage because the Ce-144 atom number density reflects the more recent exposure of the fuel in a reactor.

Zr-95($T_{1/2}$=64 days) is useful only for very short irradiations because of its half-life.

In this paper Cs-137 was selected as a burnup monitor because spent PWR fuel had a relatively long cooling time, i.e., about 8 years. The burnup was evaluated by measuring Nd-148 which was chemically separated from other fission products [5].

An empirical relation between the gamma-ray intensity of Cs-137 and the burnup was derived from the experimental results for the burnup evaluation of spent fuel.

II. Experimental

The experimental procedure for burnup evaluation is shown in Fig. 1. The gamma-ray of Cs-137 was analyzed by gamma-ray spectrometry and the amount of Nd-148 in the sample was mass spectrometry.

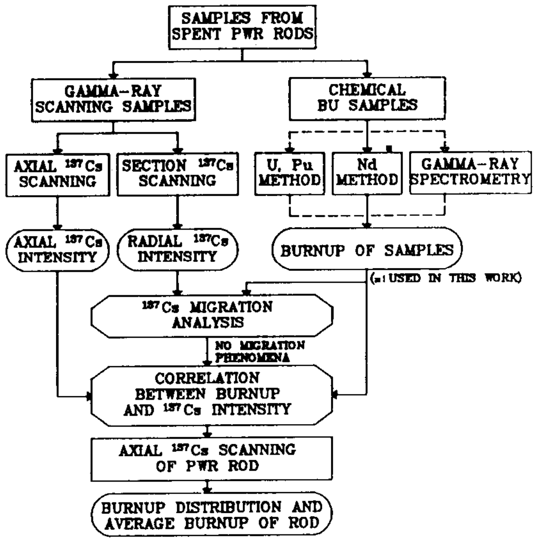

Fig. 1. Experimental Procedure for Burnup Evaluation of Spent PWR Fuel by Measuring Radioactive Fission Product Cs-137

The gamma-ray spectrum obtained from the spent fuel with a cooling time of about 8 years is shown in Fig. 2. The radioisotopes with intensive gamma-ray peaks are identified as Cs-134, Cs-137, Ce-144 and Eu-154. Cs-134 is primarily produced by the neutron activation of Cs-133 that comes from I-133 through Xe-133, and Eu-154 is essentially a product not from the fissioning of U-235 and Pu-239 but from the multiple neutron absorption of lower mass fission products. The radioisotopes having relatively short half-life, such as Zr-95 and Ru-106, are not observed in that spectrum.

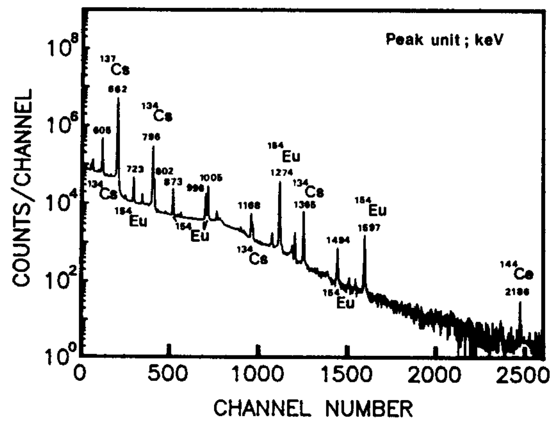

Fig. 2. Gamma-ray Spectrum Obtained from Spent PWR Fuel with a Cooling Time of about 8 Years.

In this study the spent PWR fuel rod was scanned axially by measuring 662 keV gamma-ray intensity of Cs-137 using a high purity Ge detector associated with a collimator having a 0.5 mm slit in width. In a multi-channel scaling mode, the only 662 keV signals passed through a single channel analyzer were selectively collected in a multi-channel analyzer. The rod was axially scanned from the bottom end to the spacer of the rod by using a stepping motor in order to obtain the relative distribution of Cs-137 intensity.

Two or three positions having axially flat distributions in the Cs-137 gamma-ray intensity were selected for destructive analyses. Each position was cut cross-sectionally by a micro cutter for taking two samples ; one for the sectional gamma-ray scanning and the other adjacent one for the burnup measurement by the Nd-148 method. The sample of about 3 mm thick for the sectional gamma-ray scanning was mounted with phenolic powder to protect it from breaking away and to handle it easily. The pellet for the burnup measurement by the Nd-148 method was separated totally from the cladding and dissolved with nitric acid in hot cells, and then was diluted in order to handle safely outside the biological shielding cells.

To measure the radial distribution of Cs-137 and to check the migration effect and the crack defect in the mounted sample, that may result in uncertainties in the gamma-ray measurements, the sectional gamma-ray scanning of the sample was carried out by using a 0.5×0.5 mm collimator. The cross sectional surface of a sample was divided into about 120 divisions and the collimator center could view each division by a programmable stepping motor controller.

The variation of gamma-ray intensity due to the difference from the standardized thickness of sample, 3 mm, was compensated by calculation in consideration of the attenuation of gamma-rays inside the UO_2 pellet. The intensity loss by decay of gamma-ray during cooling time and irradiation time was compensated under the assumption of constant power during irradiation in a reactor.

On the other hand, the Cs-137 gamma-ray intensity of sample was measured at axial gamma-ray scanning system and the burnup of scanned sample was evaluated by measuring Nd-148 which was chemically separated from other fission products. From these experimental results, a linear relation(LR) between the Cs-137 gamma-ray intensity and the burnup was then obtained.

In order to validate the LR, the Cs-137 gamma-ray intensity of unknown sample was nondestructively measured and the burnup obtained by the LR was compared with that of the Nd-148 method.

Ⅲ. Results and Discussion

Fig. 3 shows the distribution of the relative intensity of Cs-137 as a function of relative radius of the fuel rod samples which had different burnup. The Cs-137 gamma-ray intensity increases gradually from the center to the periphery of the rod regardless of the burnup except the lowest one which shows rather flat tendency. It seems that cracks created in the pellet do not give rise to

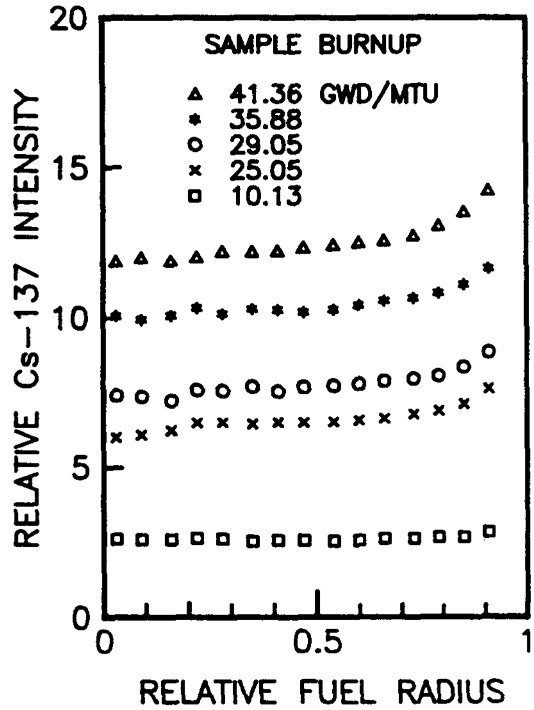

Fig. 3. Relative Cs-137 Gamma-ray Intensity as a Function of the Relative Radius of Fuel Rod Samples Which had Different Burnup

any abnormalities in the Cs-137 gamma-ray scanning.

Fig. 4 shows the linear relationship of the burnup against the normalized Cs-137 gamma-ray intensity measured by the axial gamma-ray scanning system.

The empirical equation derived from the least-squares fitting is as follows:

$$BU(GWD/MTU) = 8.688 \cdot I \cdot F + 0.488$$

Here I stands for the Cs-137 gamma-ray intensity (cps/200). The correction factor F, which generally becomes one, can be determined by taking the ratio of gamma-ray intensity of the sample used for the above equation to that measured by re-examination of the very sample when the change of experimental equipment happens or the instability of the detection system occurs.

The above relation makes it easy to determine the burnup from the results of an axial gamma-ray scanning for the spent fuel rod.

Validation test was carried out and the results are listed in Table 1. As can be seen in Table 1, the estimated burnup by gamma-ray scanning method is well agreeable with that of the Nd-148 method.

According to these results of the validation test, the burnup distribution and the average burnup of rod were determined directly by putting Cs-137 gamma-ray intensities obtained by axial scanning of rod into the empirical equation. In Fig. 5, the

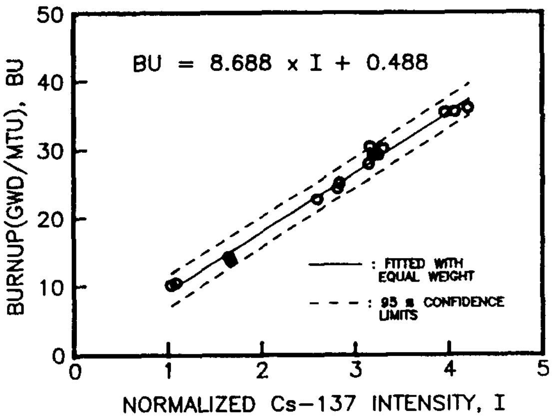

Fig. 4. Relation between the Normalized Cs-137 Gamma-ray Intensity Measured with an Axial Gamma-ray Scanning System and the Burnup

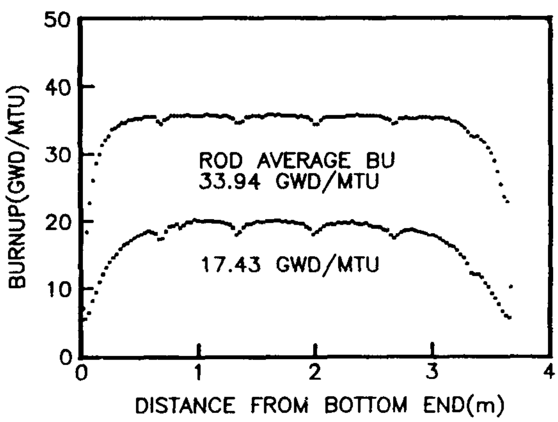

Fig. 5. Axial Burnup Distribution of Two Rods with Different Rod Average Brunup

Table 1. Comparison of Sample Burnup Measured by Gamma-ray Scanning Method with That of the Nd-148 Method

SAMPLE NO.	BURNUP (GWD/MTU)	
	ESTIMATED BY MEASURING Cs-137 GAMMA-RAY	MEASURED BY Nd-148 METHOD
1	41.01±2.42	40.88
2	40.16±2.40	40.24
3	25.77±2.18	25.70
4	25.08±2.18	24.83
5	20.08±2.20	19.82
6	5.81±2.51	5.84
7	5.62±2.52	5.27

Note : Errors refer to 2σ

rod with 34 GWD/MTU shows more flat burnup distribution than the one with 17 GWD/MTU throughout the active length except both edges.

Ⅳ. Conclusion

From the above experimental results, a conclusion is drawn up as follows :

A. The burnup is linearly related with the Cs-137 gamma-ray intensity in the range from 10 to 35 GWD/MTU ; and

B. The axial burnup profile and the average burnup of a spent PWR fuel rod is estimated directly from the results of Cs-137 gamma-ray scanning by using the empirical equation obtained in this study.

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