Determination of Plutonium Contribution to the Total Burnup of a Spent Nuclear Fuel by Mass Spectrometric Measurements of Uranium and Ruthenium

Jung Suk Kim, Young Shin Jeon, Kwang June Park, Soon Dal Park, Sun Ho Han, Yong Joon Park
Korea Atomic Energy Research Institute, 1045 Daedeok-daero, Yuseong-gu, Daejeon, 305-353, Korea,
njskim1@kaeri.re.kr

1. Introduction

In order to check the consistency of the post-irradiation analysis results, correlations between the parameters of the irradiated nuclear fuels such as the concentration of the heavy elements and fission products, ratios of their isotopes and burnup were established. These correlations can be used to identify the reactor fuels and to estimate the burnup and Pu production. A new approach was carried out with Ru isotopic ratio($^{101}Ru/^{104}Ru$) for the determination of Pu contribution to the total burnup of a spent nuclear fuel from a power reactor. The principle of this approach was based on the use of the difference in the fission yield ratios of the Ru fission products involved for the three main fissionable nuclides such as ^{235}U, ^{239}Pu, and ^{241}Pu.

In this work, to determine the contribution of Pu to the total burnup of the fuel, the following two independent methods have been applied: 1) by measuring the isotope ratios of the stable Ru fission products $^{101}Ru/^{104}Ru$, and 2) by determining the total burnup by Nd-148 method[1] and substracting partial(^{235}U) burnup[2,3], which is determined from the measured values of U isotope ratios.

2. Methods and Results

2.1 Fuel Dissolution and Sample Preparation

A spent fuel sample weighed exactly was placed in a dissolution flask of dissolution apparatus in hot cell. The fuel sample was refluxed for more than 16 hours in HNO_3(1+1) without a catalyst. This fuel solution was weighed and an aliquot was diluted with the HNO_3(1+1) with the aid of ORIGEN calculation for the estimation of nuclides content in the spent nuclear fuel.

2.2 Separation Procedure

Chemical separation of U, Pu and Nd for burnup determination was carried out for both of the unspiked and the spiked(^{233}U, ^{242}Pu and ^{150}Nd) sample solutions under the same experimental conditions in glove box without heavy shieldings[2]. The procedure using a distillation method, which is based on the volatility of RuO_4 was carried out in this work in order to separate Ru from the fuel solution[4]. The pure separation of Ru is identified by a series of preliminary experiments with the prepared solution, followed by chemical analysis of the ICP-AES and EPMA.

2.3 Determination of Isotopic Composition

The U, Pu and Nd fractions collected from the spiked and unspiked fuel samples. The Ru fraction in absorbtion solution were prepared for mass spectrometric determination. The isotopic compositions of U, Pu, Nd and Ru fractions were determined by using a mass spectrometer of the Finnigan MAT 262 type, respectively(Table 1).

2.4 Determination of Pu Contribution by Ru Isotope Method

Assuming that the total burnup of the fuel is the sum of partial contributions due to U and Pu, $F_U+F_{Pu}=100$,

$$R_{ij} = \frac{F_U Y^i_U + F_{Pu} Y^i_{Pu}}{F_U Y^j_U + F_{Pu} Y^j_{Pu}} \quad \cdots\cdots (1)$$

We finally obtain the following equation for Pu contribution, F_{Pu}, to the total burnup:

$$F_{Pu} = \frac{(R_{ij}\ Y^j_U - Y^i_U) \times 100}{Y^i_{Pu} - Y^i_U - R_{ij}(Y^j_{Pu} - Y^j_U)} \quad \cdots\cdots (2)$$

Where R_{ij} : atom ratio of ^{101}Ru to ^{104}Ru in the spent fuel sample

Y^i_U and Y^j_U : thermal fission yield of ^{101}Ru and ^{104}Ru for ^{235}U, respectively

Y^i_{Pu} and Y^j_{Pu} : weighted average thermal fission yield of ^{101}Ru and ^{104}Ru for ^{239}Pu and ^{241}Pu, respectively

Y values are calculated from the equation in reference[4] with the chemical analysis results for fissionable isotopes and their nuclear data[5]. The fission yields of Ru isotopes by thermal neutron used for calculation are shown in Table 2. Based on this equation(2), the calculated value of Pu contribution(%), F_{Pu} are presented in Table 3. In this case, it is ignored that the Pu contribution by the fast fission of ^{238}U.

Table 1. Isotopic Compositions of the Ru separated from a spent nuclear fuel sample

	Atom%					
	^{99}Ru	^{100}Ru	^{101}Ru	^{102}Ru	^{104}Ru	^{106}Ru
E*	0.312	9.939	32.45	33.91	23.39	-
C+	0.002	7.027	33.15	35.10	24.30	0.418

*measured experimentally by chemical method
+calculated by ORIGEN-ARP code for rod average

Table 2. Fission yield(%) of Ru Isotopes by thermal neutron(t)

Isotope	^{235}U(t)	^{239}Pu(t)	^{241}Pu(t)	W.A.F.Y*
^{101}Ru	5.1729	6.0192	6.2319	6.0792
^{104}Ru	1.8808	6.0948	7.1795	6.4006

*weighted average fission yield for ^{239}Pu and ^{241}Pu

2.5 Determination of Pu contribution by burnup difference method

Table 2 shows the Pu contribution(%) value by the burnup difference method, that is, {[(total burnup-^{235}U burnup)/total burnup] x100}. The total burnup of the fuel sample, F_t was determined from the analysis of ^{148}Nd, U and Pu(Nd-148 method) by the following equation:

$$F_t = \frac{N/Y}{N/Y+N(U)+N(Pu)} \times 100 \quad \cdots\cdots (3)$$

Where F_t : total burnup in atom% fission

N : number of atoms of the monitor Nd isotope in the spent fuel solution,

Y : effective fission yield of the monitor Nd isotope from the fissile elements,

N(U), N(Pu) : number of U and Pu atoms in the spent fuel solution, respectively

For the calculation of U contribution to the total burnup of the fuel sample, the fractional burnup due to a fission of ^{235}U, F_5 was calculated by the equation shown in references[2,3].

The results obtained by the Ru isotope ratio and the burnup difference method for determination of Pu contribution to the total burnup, are in a good agreement, within a deviation of 4.41%(Table 3).

Table 3. Determination of Pu contribution to the total burnup of a spent nuclear fuel

No. of Fissions per g-fuel	Total Burnup (GWD/MtU)	Pu Contribution(%)	
		Ru Isotope Ratio	Burnup Difference
1.330×10^{20}	57.5±0.2	47.83	45.72

3. Conclusions

The U and Ru isotope patterns provide information on the real irradiation characteristics which are necessary for evaluating a fuel's performance in a reactor. A comparison of the Pu contribution values determined independently provides a promising way to check on the validity of the results.

4. References

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