Radial Distribution of Isotope Ratios in Spent Nuclear Fuels with Different Burnups

Yeong-Keong Ha*, Jung Suck Kim, Young Shin Jeon, Hang Seok Seo, Kyuseok Song
Korea Atomic Energy Research Institute, 1045 Daeduckdaero, Yuseong-gu, Daejeon, 305-353, Korea
**Corresponding author: nykha@kaeri.re.kr*

1. Introduction

During nuclear power generation, some isotopes are produced by the fission process causing the compositional changes in a nuclear fuel. The abundances of some isotopes provide information about fuel burnup. Direct analysis of fission products in a spent fuel by laser ablation inductively coupled plasma mass spectrometric analysis (LA-ICP-MS) was carried out to provide local chemical distributions of fission products from the core to the periphery of a fuel pellet. To have a better understanding of local chemical properties, spent nuclear fuels with different burnups from 30,000 to 60,000 MWd/MtU were chosen as sample specimens. The local burn up based on isotopic measurements of Np, Am, Cm by LA-ICP-MS are presented.

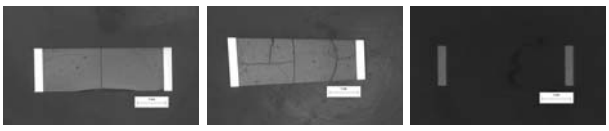
2. Experimental

2.1 Instruments

ICP-MS has been used as powerful analytical tool for the determination of isotope ratios at the trace level due to its sensitivity and accuracy. By combining the laser ablation technique with ICP-MS, micro sampling of a heterogeneous sold sample was achieved [1]. For a highly radioactive sample analysis, we built a shielded LA system in order to avoid the dose to an operator and details of this system were described in our previous paper [1].

2.2 Specimen preparation

Spent fuels originating from the Yeonggwang-2 nuclear reactor were chosen as the test specimens. For comparison, two specimens were taken from the same fuel rod at axial positions of 180 mm (P14P17-1) and 3340 mm (P14P17-2) from the bottom of the fuel rod, and a specimen was taken from another fuel rod (P11Q01). The spent fuels were cut along the diameter of the pellet including the cladding by 3 mm (width) x 1 mm (thickness) and embedded in epoxy resin and then polished well in the PIE facility in KAERI (Fig. 1).



(a) (b) (c)

Fig. 1. Prepared spent nuclear fuel specimens: (a) P14P17-1, (b) P14P17-2, (c) P11Q01.

3. Results and Discussions

3.1 Burnup Determination by Nd Method

For the measurement of fuel burnup, two pieces (P14P17-1, P14P17-2) of P14P17 spent fuel with 48.7 GWd/MtU rod average burnup and two pieces (P11Q01-2, P11Q01-5) of P11Q01 spent fuel with 55.8 GWd/MtU rod average burnup were analyzed by chemical dissolution method. Dissolution of the samples were carried out by HNO_3(1:1) and by c-HNO_3 at 90 ℃ for 8 hours individually. After dissolution, the dissolved solutions were diluted by HNO_3(1:1). Spent fuel samples with and without spike (^{233}U, ^{242}Pu, ^{150}Nd) addition were prepared and then U, Pu and Nd were separated by ion exchange chromatography (Biorad AG 1x4 and 1x8). Each fraction of U, Pu and Nd were collected from the sample solution and their isotopic compositions were measured by mass spectrometry. The details of separation process are described in elsewhere [2, 3]. The isotope compositions were measured using thermal ionization mass spectrometer (TIMS) and the calculated burnups are listed in Table 1.

Table 1. Total burnup determined by the Nd isotope monitor methods for the PWR fuel samples (unit: GWd/MtU).

Sample	Nd-148	Nd-(145+146)	Nd-Total*
P14P17-1	33.33±1.04	34.16±1.07	33.52±1.05
P14P17-2	40.97±1.28	41.96±1.31	41.37±1.29
P11Q01-2	58.21±1.82	60.30±1.88	58.65±1.83
P11Q01-5	57.00±1.78	58.81±1.84	57.82±1.81

* 143+144+145+146+148+150

Both samples of P14P17 revealed lower burnups than rod average burnup of 48.7 GWd/MtU, and P11Q01 samples revealed higher burnups than the rod average burnup of 55.8 GWd/MtU calculated from ORIGEN2.

3.2 Distribution of Isotope Ratio

Among the fission products, the isotope ratios of ^{236}U, ^{239}Pu, ^{240}Pu ^{241}Pu, ^{237}Np, ^{243}Am, ^{244}Cm, ^{100}Mo, ^{137}Cs, and ^{144}Nd in a spent fuel specimen were measured. The peak ratios to that of ^{235}U were used to study the radial isotopic distributions of uranium, plutonium, actinides and other fission products. Sampling was carried out with 500 μm intervals around core, and then with

reduced intervals of 200~300 μm in the middle, and finally with 100 μm interval near the rim zone.

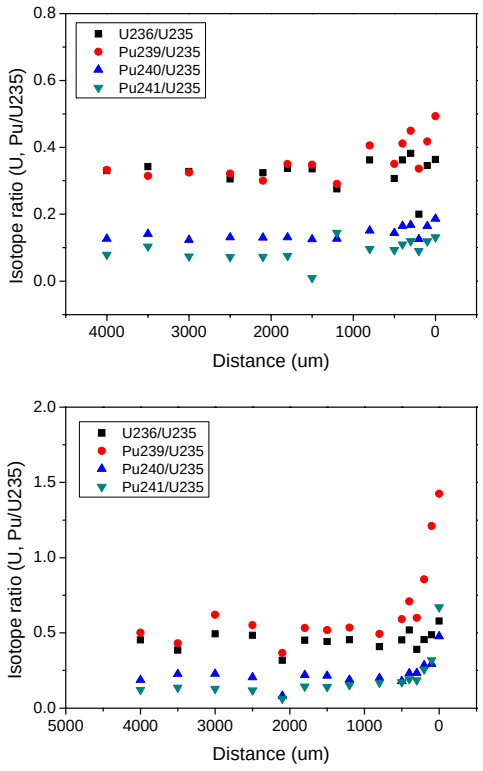


Fig. 2. Distribution of ^{236}U, ^{239}Pu, ^{240}Pu and ^{241}Pu to ^{235}U: above) 33,7 GWd/MtU, below) 41.4 GWd/MtU

Fig. 2 shows the isotope ratios of ^{236}U and ^{239}Pu, ^{240}Pu, ^{241}Pu to ^{235}U from the core to the rim of the fuel pellets with 33.7 and 41.4 GWd/MtU. As can be observed, ^{236}U (formed by neutron capture of ^{235}U) to ^{235}U ratio is almost constant, while those of Pu isotopes increase significantly at the rim zone as the burnup increase. The higher Pu isotopes are known to be a result of successive neutron capture reactions by ^{238}U at the pellet periphery. Distribution of actinides (^{237}Np, ^{243}Am, ^{244}Cm) and some other fission products (^{100}Mo, ^{137}Cs, ^{144}Nd) was also observed. Local fuel burnup values based on the measured isotope ratios for selected elements were calculated by applying ORIGEN2 code values.

3.3 Reproducibility of Isotope Ratio

Since a spent fuel reference material does not exist, the reliability of the measured data was determined by the reproducibility of the isotope ratios measured at the same radial position. The relative standard deviations (RSD) of the measurements were in the range between 5% to 20% depending on the element and the specimen burnup.

4. Conclusions

The radiation shielded LA-ICP-MS was employed to measure the isotopic distribution of U, Pu and other fission products in a spent nuclear fuel discharged from Yeonggwang-2 power plant. It was observed that the isotopic ratio of ^{236}U to ^{235}U is almost constant from core to rim of the fuel pellet and that of Pu increased significantly near the pellet periphery due to the neutron capture reactions by ^{238}U (so-called rim effect). Changes in the isotopic distribution of MA and other fission products were observed at different specimen burnups. The obtained data provide local burnup profiles, and are useful for better understanding of irradiation behaviors of a high burnup spent nuclear fuel.

Acknowledgements

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