

Transuranium Inventory in High Burnup Spent Fuel Samples by An Alpha Spectrometry

Kihsoo Joe, Byung-Chul Song, Young-Bok Kim, Sun-Ho Han, Young-Shin Jeon, Euo-Chang Jung
 and Kyuseok Song

Korea Atomic Energy Research Institute 150 Duckjindong, Yusong, Taejon, Korea 305-353 e-mail :
ksjoe@kaeri.re.kr

1. Introduction

Determination of actinide elements in spent nuclear fuels is important for a source term evaluation, burnup credit evaluation and actinide inventory. In this study, transuranic elements such as Pu, Np, Am and Cm were determined by an alpha spectrometry in high burnup spent nuclear fuel samples ranging from 36 to 55 GWD/MtU as a burnup. The used separation method was an anion exchange chromatography[1] and/or a diethylhexyl phosphoric acid(HDEHP) extraction chromatography[2] as shown in Fig 1. The separated elements were electrodeposited. Alpha spectrometry was applied for ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{237}Np , ^{241}Am , ^{244}Cm and ^{242}Cm using ^{242}Pu and ^{243}Am as the tracers, respectively. Especially, for ^{237}Np , a spike addition technique was adopted using ^{239}Np as a spike because these spent nuclear fuel samples contain a considerable amount of ^{239}Np [3,4].

The determined contents of transuranic elements were compared with those calculated by the Origen-2 code[5].

2. Experimental

Six high burnup PWR spent fuel samples(35.6 ~ 53.9 MWd/MtU) with different cooling times of about 2.2 ~ 6.3 years were take from U1Jin unit 2 and Youngkwang unit 4. These samples were dissolved with (1+1) HNO_3 in a hot cell and the solutions were diluted to an appropriate concentrations. An optimum sample size was estimated by calculating the amount of each nuclide according to a burnup using the Origen-2 code. An appropriate amount of a sample was taken and loaded onto an anion exchange column after a sample pretreatment. Prior to a sample loading, the oxidation states of the elements were controlled to be Pu(IV) and Np(IV) by a $\text{NH}_2\text{OH}\cdot\text{HCl}$ reductant. Pu was eluted by a reduction to Pu(III) with a 9 M HCl-0.1M HI eluent after a sample loading in a 9M HCl-0.1M HNO_3 medium in which Am and Cm including fission products were eluted without an adsorption onto the anion exchange column. Np was eluted with a 4 M HCl eluent after a Pu elution. Am and Cm were separated from the fission products by a HDEHP extraction chromatography[2] using a DTPA-lactic acid eluent. The separated elements were determined by alpha spectrometry after an electroduction. For ^{237}Np , ^{239}Np was added into the sample as a spike and it was determined by an isotope dilution alpha(gamma)

spectrometry.

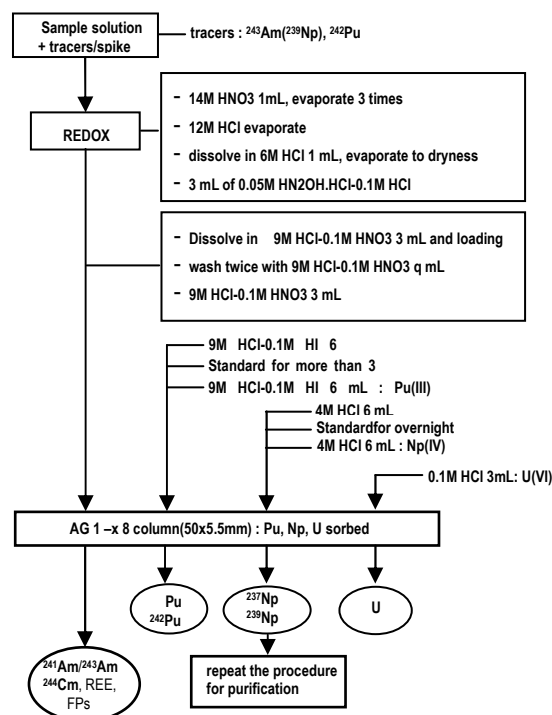


Fig 1. Sequential separation of transuranic elements in spent nuclear fuel sample solutions

3. Results and discussion

3.1 Determination of Pu isotopes

The Pu isotopes were determined by an alpha spectrometry using ^{242}Pu as a tracer. The alpha activities of ^{238}Pu and $^{239+240}\text{Pu}$ were measured at 5.5 MeV and 5.16 MeV, respectively. Each activity of ^{239}Pu and ^{240}Pu was obtained by a mass spectrometry for a Pu fraction. The measured values by this method were 0.274 ~ 0.584 $\mu\text{g } ^{238}\text{Pu}/\text{mgU}$, 6.0 ~ 7.05 $\mu\text{g } ^{239}\text{Pu}/\text{mgU}$ and 2.57 ~ 3.55 $\mu\text{g } ^{240}\text{Pu}/\text{mgU}$, respectively. The measured values were also compared with those by the Origen-2 code. On the whole, the measured values showed higher values than the calculations as follows: 27.7 ~ 50.3% for ^{238}Pu , -8.0 ~ 17.2% for ^{239}Pu and 7.9 ~ 21.8% for ^{240}Pu . In Fig 2 a correlation between the measurement and the calculation for ^{239}Pu was found as a representative one.

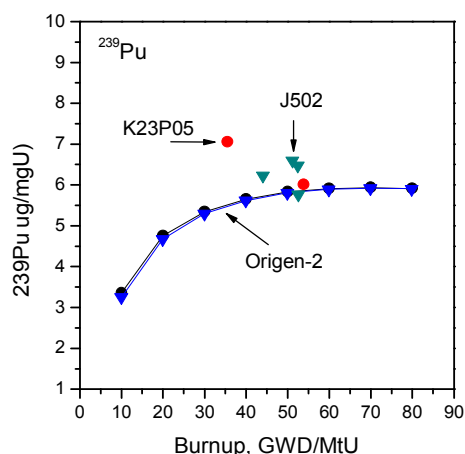


Fig 2. Correlation of ^{239}Pu between measurement and calculation in spent fuels. K23P05 : cooling time 2302 day, J502D16/J502A14 : cooling time 788 day

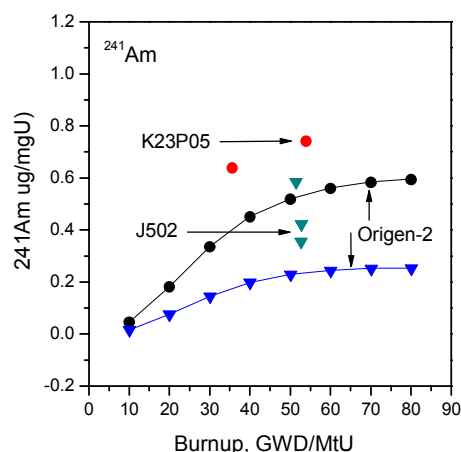


Fig 3. Correlation between measurement and calculation for ^{241}Am

3.2 Determination of ^{237}Np

A spike addition technique was adopted for the determination of ^{237}Np using ^{239}Np as a spike. About 60 Bq of ^{239}Np was added into one sample (“a spiked sample”). The other sample was a “sample” in which a spike was not added. Two “measurements” were performed separately for each anion exchange column for one sample. The ^{237}Np was measured by an alpha spectrometry (4.79 MeV) and ^{239}Np by a gamma spectrometry (228.07KeV/277.86KeV) after an electrodeposition. The content of ^{237}Np was calculated by an isotope dilution alpha(gamma) spectrometry. The content of ^{237}Np was in the range from 0.35 to 0.89 $\mu\text{g}/\text{mgU}$ and showed the lower values by -16 ~ -66% when compared with those by the calculation.

3.2 Determination of Am and Cm

For the determination of Am and Cm, a small amount of a sample (~ 0.1 μg of U) was taken. About 15 Bq of ^{243}Am as a tracer was added into a sample solution.

The sample solution was loaded onto the HDEHP extraction column. The Am and Cm were eluted together with 6 mL of 0.05 M DTPA-0.5 M LA with successive additions of 3 mL each. Then, the lanthanides as well as Mo and Zr were eluted with 5 mL of 6 M HNO_3 . But, this step was omitted in this work.

The separated nuclides were determined by an alpha spectrometry and the contents of the nuclides were compared with those by the Origen-2 code. The contents of the nuclides were 0.36 ~ 1.09 $\mu\text{g}/\text{mgU}$ for ^{241}Am , 6×10^{-6} ~ 1.9×10^{-3} $\mu\text{g}/\text{mgU}$ for ^{242}Cm and 0.05 ~ 0.3 μg for ^{244}Cm , respectively. The measured values were much higher than the calculations by 22.6 ~ 78.9 % for ^{241}Am , 11.6 ~ 59.3% for ^{242}Cm and 63.0 ~ 82.8% for ^{244}Cm , respectively. The correlation between the measurements and the calculations for ^{241}Am is shown in Fig 3.

3. Conclusion

The contents of transuranic elements as a source term data for high burnup spent fuel samples was determined. A comparison between the calculation and the measurement showed that this was lower than that for ^{237}Np , while for the other nuclides this was higher on the whole. In the future more data will be required for a code verification related to high burnup spent fuels.

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