*Transactions of the Korean Nuclear Society Autumn Meeting*
*PyeongChang, Korea, October 30-31, 2008*

# Determination of Pu, Am and Cm in High Burnup Spent Fuel Samples by an Alpha Spectrometry

Do-Yang Kim, Kihsoo Joe, Young-Shin Jeon, Sun-Ho Han, Euo-Chang Jung and Kyuseok Song
*Korea Atomic Energy Research Institute 150 Duckjindong, Yusung, Taejon, Korea 305-353 e-mail : ndykim@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, Am and Cm in high burnup spent nuclear fuel samples ranging from 55 to 62 GWD/MtU as a burnup were determined by an alpha spectrometry. 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 [3]. The alpha activities of the nuclides such as $^{238}$Pu, $^{239}$Pu , $^{240}$Pu, $^{241}$Am, $^{244}$Cm and $^{242}$Cm were determined by using $^{242}$Pu and $^{243}$Am as the tracers, respectively. The determined contents of transuranic elements were compared with those calculated by the ORIGEN-2 code [4].

## 2. Experimental

Three high burnup PWR spent fuel samples(55 ~ 62 MWd/MtU) with different cooling times of about 2.6 ~ 7.1 years were take from UlJin unit 2 and Youngkwang unit 2. 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) by a $NH_2OH.HCl$ 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 (Fig 1). Am and Cm were separated from the fission products by a HDEHP extraction chromatography[2] using a DTPA-latic acid eluent. The separated elements were determined by an alpha spectrometry after an electrodeposition.

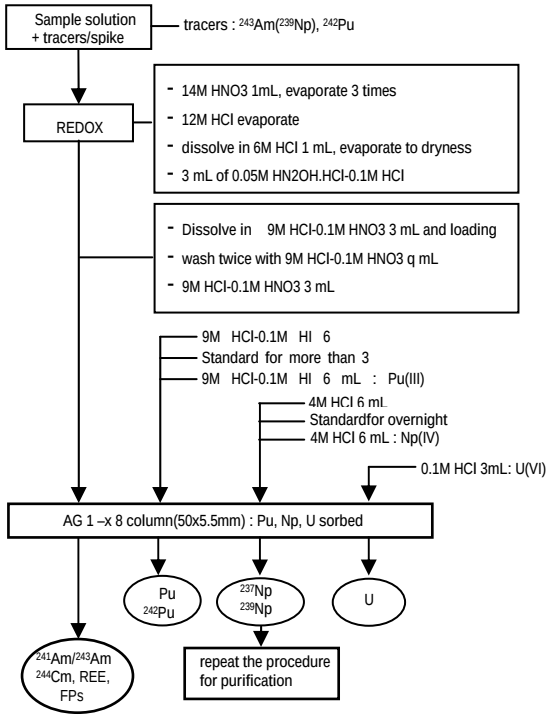



Fig 1. Sequential separation of transuranic elements

## 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 $^{239+240}$Pu were measured at 5.5 MeV and 5.16 MeV, respectively (Fig. 2). 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 0.487 ~ 0.535 μg/mgU for $^{238}$Pu, 5.333 ~ 5.780 μg /mgU for $^{239}$Pu and 2.776 ~ 3.407 μg/mgU for $^{240}$Pu, respectively, as shown in Table 1. The measured values were also compared with those by the ORIGEN-2 code.

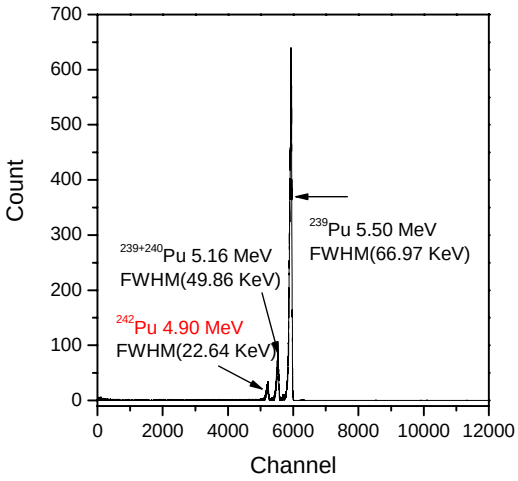
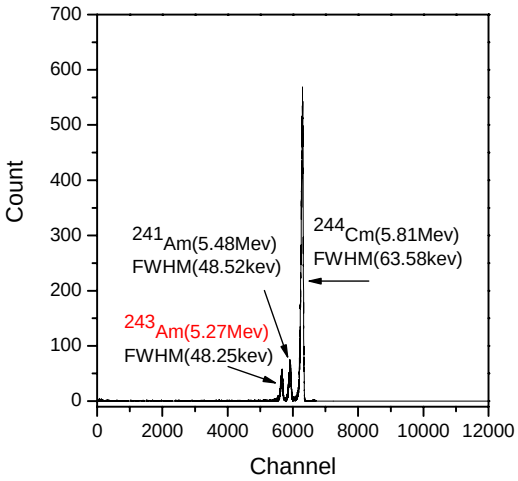



Fig. 2. Alpha spectrum of the Pu isotopes




Fig, 3. Alpha spectrum of Am and Cm

*3.2 Determination of Am and Cm*

For the determination of Am and Cm, a small amount of a sample(~ 0.1 g of U) was taken. About 15 Bq of $^{243}$Am as a tracer was added into a sample solution. The sample solution eluted from the anion exchange column was loaded onto the HDEHP extraction column after sample pretreatment by nitric acid. 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. The separated nuclides were determined by an alpha spectrometry (Fig. 3). The measured values were 0.3692 ~ 0.6088 for $^{241}$Am, 5.3x10$^{-6}$ ~ 6x10$^{-4}$ for $^{242}$Cm and 0.1895 ~ 0.2499 μg/mgU for $^{244}$Cm, respectively, as shown in Table 1. The contents of the nuclides were compared with those by the ORIGEN-2 code.

Table 1. Amount of transuranium elements(μg/mgU)

| Nuclides | SF-1 | SF-2 | SF-3 |
|---|---|---|---|
| $^{238}$Pu | 0.5351 | 0.5058 | 0.4867 |
| $^{239}$Pu | 5.6440 | 5.7800 | 5.5334 |
| $^{240}$Pu | 3.4068 | 3.0224 | 2.7762 |
| $^{241}$Am | 0.6088 | 0.4717 | 0.3692 |
| $^{242}$Cm | 5.3E-06 | 0.0006 | 0.0007 |
| $^{244}$Cm | 0.2356 | 0.1895 | 0.2499 |

## 4. Conclusion

The contents of transuranic elements in high burnup spent fuel samples were determined. A comparison between the measurement and the calculation by the ORIGEN-2 code was performed. In the future more data will be needed for a code verification related to high burnup spent fuels.

## REFERENCES

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