

DETERMINATION OF THE TRANSURANIC ELEMENTS INVENTORY IN HIGH BURNUP PWR SPENT FUEL SAMPLES BY ALPHA SPECTROMETRY-II

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The contents of transuranic elements (^{237}Np , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Am , ^{244}Cm , and ^{242}Cm) in high-burnup spent fuel samples (35.6~53.9 GWd/MtU) were determined by alpha spectrometry. Anion exchange chromatography and diethylhexyl phosphoric acid extraction chromatography were applied for the separation of these elements from the uranium matrix.

The measured values of the nuclides were compared with ORIGEN-2 calculations. For plutonium, the measurements were higher than the calculations by about 2.6~32.7% on average according to each isotope, and those for americium and curium were also higher by about 35.9~63.1%. However, for ^{237}Np , the measurements were lower by about 52% on average for the samples.

KEYWORDS : Inventory of Transuranic Elements, High-Burnup Spent Fuels, Alpha Spectrometry, Isotopic Analysis, Spike Addition

1. INTRODUCTION

Transuranic element (such as Np, Pu, Am, and Cm) content has been determined for a spent fuel chemical characterization [1,2], an evaluation of a burnup credit [3,4,5,6], and tracing the origin of nuclear materials [7]. For example, if a sample of depleted uranium (DU) contains trace levels of ^{236}U , ^{237}Np , ^{239}Pu , ^{240}Pu , and ^{241}Am , it is likely that it originated from the chemical reprocessing of spent fuel, as analyses have shown. This was done through measuring the transuranic element (^{239}Pu , ^{240}Pu , ^{241}Am and ^{237}Np) content through alpha spectrometry and inductively coupled plasma-mass spectrometry (ICP-MS) using ^{242}Pu and ^{243}Am as tracers [8]. Additionally, measurement of the transuranic element content in spent nuclear fuel is also used for a code verification that predicts nuclides produced or decayed during a neutron irradiation.

A number of separation methods have been used to determine the transuranic element content in spent nuclear fuel, such as anion exchange chromatography using nitric acid [9] or hydrochloric acid [6, 10] eluents. For the purification of Am from UO_2 , a tri-n-octyl amine (TNOA) column followed by a diethylhexyl phosphoric acid (HDEHP) column was used [8]. At pH 2.4 in this system, Am is highly and selectively adsorbed onto these columns. But at very low pH (1 M HNO_3), Am can be eluted from these columns.

It has been reported that transuranic elements (Np, Pu, Am, and Cm) in UO_2 and mixed oxide (MOX) fuel solutions have been separated and identified by an ion chromatography (IC)-ICP-MS system using a cation exchange column (CS10) and 2,3-diaminopropionic acid monohydrate (DAP) as eluent [1]. This system demonstrated good elemental resolution.

A widely used technique for the determination of actinide elements in environmental and radioactive samples is electroplating followed by alpha spectrometry. This choice is due to its low cost in comparison to other isotopic techniques such as ICP-MS [11, 12, 13]. Tracers are also required for determination of transuranic element content in environmental and spent fuel samples. Special attention is needed in the selection of appropriate tracers according to the elements employed in the determination methods. In general, ^{243}Am , ^{248}Cm , and ^{242}Pu tracers are recommended for determining Am, Cm and Pu isotope content, respectively, in these samples [10], and for the determination of ^{237}Np , tracers such as ^{235}Np or ^{236}Np can be used [14, 15].

As ^{248}Cm , ^{235}Np , and ^{236}Np were not available in our laboratory. So, in this work, ^{243}Am and ^{239}Np were used as a tracer and a spike, respectively. Since ^{239}Np , which emits low gamma energies, has a very short half-life (2.35 d), it needs regular weekly preparation and standardization which is time consuming and expensive. Therefore, in

this work, a standard ²⁴³Am solution was used directly for ²³⁹Np as a spike because it was already equilibrated with ²³⁹Np in the standard solution.

In this study, high-burnup pressurized water reactor (PWR) spent fuel samples were analyzed to determine the inventory of the transuranic elements. Anion exchange chromatography using a hydrochloric acid eluent [9, 10] and diethylhexyl phosphoric acid (HDEHP) extraction chromatography [16] were applied in serious or not. After electroplating, the activities of the separated elements were measured by alpha spectrometry. Finally, the measured contents were compared with ORIGEN-2 calculations.

2. EXPERIMENTS

2.1 Reagents and Apparatus

The anion exchanger (AGMP-1 x 8, 100-200 mesh size) used in this work was manufactured by Bio-Rad Laboratories, USA. Diethylhexyl phosphoric acid (>95%, Aldrich Chem. Co, USA) was used as the adsorbent, and diethylenetriaminetetraacetic acid (DTPA) (97%, Aldrich Chem. Co, USA), and lactic acid (LA) (85-90%, Tedia Co, USA) were used as eluents for the separation of Am and Cm. Amberlite XAD-XVI, styrenedivinylbenzene (SDVB) copolymer, used as a support material for the adsorbent was obtained from the Merck Company of Germany.

The standard solutions of ²⁴³Am (37.52 KBq ±3%, 4.961 g-soln, Nov.1 1991) and ²⁴²Pu (25.67 Bq/g, ±0.72%, 5.5 g-soln, Aug. 1, 1999) as the tracers were obtained from the North American Scientific Inc (Model Cal 3000) and NBL (CRM), USA, respectively. Standard solutions of ²⁴¹Am (417 KBq, ±2.35%, 5.037 g-soln, Aug. 1, 1994), ²⁴⁴Cm (370 KBq, ±1.67%, 5.297 g-soln, Sep. 1, 1994), and ²³⁹Pu (41.59 KBq, ±4.01%, 5.19 g-soln, Nov. 1, 1995) were obtained from the North American Scientific Inc., USA. The standard ²³⁷Np solution (832 KBq/g, ±3%, 1.066 g-soln) was obtained from the Damri Company (CEA), France. A standard solution of ¹⁵²Eu (45700 Bq/mL, January 1, 1991, code no EFY.64, Amersham Co, UK)

was used for gamma spectroscopy energy and efficiency calibrations. NaHSO₄ (extra pure, Junsei Co., Japan) and Na₂SO₄ (GR, Merck Co., Germany) were used as an electrolyte for the electroplating. The standard mixed alpha source (²⁴¹Am/²⁴⁴Cm/²³⁹Pu~1, 3.28x10⁵ dpm, July 16, 1986) used for the alpha spectrometry came from Amersham Co., UK.

The alpha (silicon surface detector of 300 mm²) and gamma spectrometers were the products of EG&G ORTEC, USA. Total alpha activity was measured by a Gas Proportional Counting System (LB 5100, Tennelec Co, USA). The mass spectrometer used for ²³⁹Pu/²⁴⁰Pu ratio measurements was the Finnigan Mat 262 type (Finnigan Mat Company of Germany). The electroplating system consisted of a polyethylene cell (inner diameter 20 mm x height 80 mm), a planchet disc of stainless steel (diameter 1 inch x thickness 0.5 mm), and a Pt electrode (diameter 1 mm x length 100 mm).

2.2 Sample Preparation

Six PWR spent nuclear fuel samples with burnups ranging from 35.6-53.9 GWd/MtU were obtained from the nuclear power plants in Korea (Table 1). The specimens were taken from different positions of the fuel pins from middle to upper parts and the fuel assemblies including sample rods were moved from out-side to inside and to out-side again of the reactor core during irradiation. The cooling times of the samples were ranged from 2.1-6.3 years. A small piece of specimen (≈0.5g) was dissolved with (1+1) nitric acid under a reflux condenser in a hot cell. The sample solutions were pneumatically transferred to a steel box after being diluted to an appropriate concentration. Separation of the transuranic elements was performed inside a fume hood.

2.3 Chromatography for the Transuranic Elements

2.3.1 Column Preparations

Column (A) was a polyethylene column (inner diameter 5.5 mm x height 50 mm) filled with anion exchanger (AGMP-1 x 8, 100-200 mesh size), and column (B) was a

Table 1. Sample History used for the Determination of Transuranic Elements

Specimen	²³⁵ U (ini.wt%)	GWd/MtU	Cooling time (d)	Location (mm from bottom)
SF-1	4.2	53.865	2302	2444
SF-2	4.2	35.557	2313	3442
SF-3	4.49	52.525	775	1839
SF-4	4.49	44.155	788	3137
SF-5	4.49	52.688	801	1607
SF-6	4.49	51.288	807	3147

polyethylene column (inner diameter 5.5 mm x height 50 mm) filled with an adsorbent (HDEHP coated onto SDVB copolymer, 100-200 mesh size: 2.1 mmole HDEHP/g-adsorbent).

2.3.2 Separation and Determination of Pu

A small amount of sample solution equivalent to approximately 0.1 µg of U (≈20 Bq ²³⁸Pu based on an estimation by ORIGEN-2 for spent fuel of 50 GWD/MtU) [17,18] was taken, and a 2.258 Bq ²⁴²Pu yield tracer was added to the solution. The sample solution was treated with nitric acid and hydrochloric acid by evaporation on a hot plate. The residue was dissolved in 3 mL of 0.05 M NH₂OH.HCl-0.1 M HCl, and left overnight to obtain Pu(IV) and Np(IV). Finally, a sample solution in a medium of 9 M HCl-0.1 M HNO₃ was loaded onto column (A). The column was washed with 9 M HCl-0.1 M HNO₃, and the procedure outlined in Figure 1 was then followed. The Pu was eluted by reduction to Pu(III) with a 12 mL 9 M HCl-0.1 M HI solution in 6 mL increments and ≈2 hour intervals to reduce the Pu(IV) or Pu(VI) to Pu(III). The eluted Pu was electroplated in a sodium hydrogen sulfate-sodium sulfate electrolyte, and the activities of the Pu isotopes were then measured by alpha spectrometry.

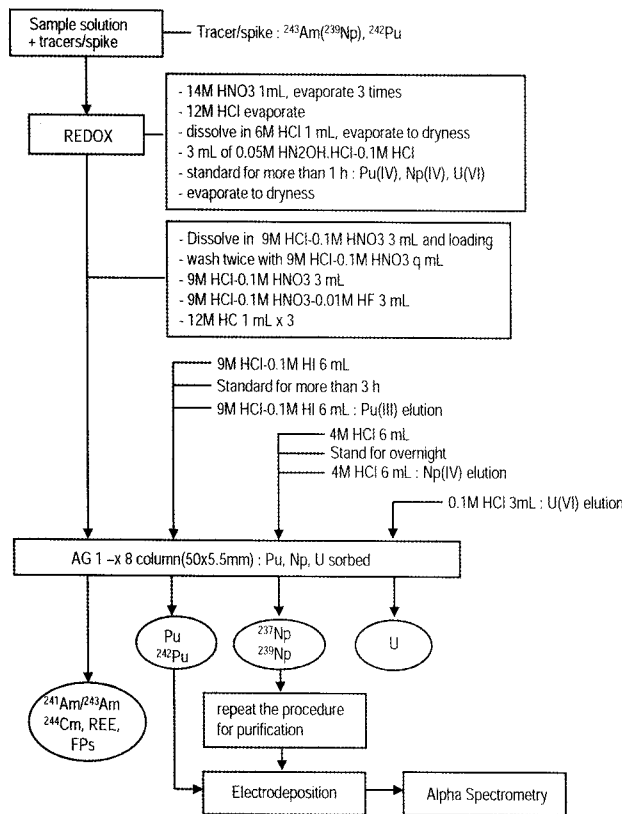


Fig. 1. Separation Scheme for Transuranic Elements in Spent Nuclear Fuel Samples

2.3.3 Separation and Determination of Am and Cm

A small amount of sample solution equivalent to approximately 0.1 µg of U (≈10 Bq ²⁴¹Am and ≈20 Bq ²⁴⁴Cm based on the same estimation as in Pu) was taken, and a 15 Bq ²⁴³Am yield tracer was added to the solution. The sample treatment and anion exchange chromatography were performed with the same procedure outlined above. An effluent (at a sample loading step) containing Am, Cm, lanthanides, and other fission products, which were not retained on the anion exchange column during the Pu adsorption, was dried on a hot plate and then transformed to nitrate salt through nitric acid treatments. The sample solution was finally loaded onto HDEHP column (B) after the medium of the solution was changed to 0.1 M HNO₃ as reported in previous work [18]. Americium and curium were eluted together from the column with 6 mL of 0.05 M DTPA-0.5 M lactic acid in successive additions of 3 mL each after a number of washing steps through column (B). Fractions of Am and Cm, including organic compounds as eluents, were heated to high temperature on a hot plate in order to decompose them through carbonization. As a next step, the carbon component was completely oxidized through a number of c-HNO₃ and c-HClO₄ treatments until the solution became clear.

This solution was again dried, and nitrate salts were obtained by c-HNO₃ treatments. Electroplating of the Am and Cm was performed with the same procedure used for the Pu. The activities of Am and Cm were then measured by alpha spectrometry just as in the case for Pu.

2.3.4 Separation and Determination of Np

For the determination of ²³⁷Np, a larger sample size equivalent to ≈100 µg U (≈2 Bq ²³⁷Np per sample) was taken due to the low specific activity of ²³⁷Np resulting from its long half life of 2.14x10⁶ y. An isotope dilution method [19] was adopted by making two measurements for one sample (“sample” and “spiked sample”) using ²³⁹Np as a spike. Approximately 60 Bq of ²³⁹Np was added to the “sample” solution (changed to “spiked sample”). The sample pretreatment was the same as that for Pu. The two samples were applied separately to each anion exchange column (A). The Np was eluted with 12 mL 4 M HCl eluent after a Pu elution (Fig. 1). The activities of ²³⁷Np and ²³⁹Np were measured by alpha and gamma spectrometry, respectively, after electroplating the two nuclides onto the same planchet. The ²³⁷Np content was calculated using the ²³⁷Np/²³⁹Np ratios in the “sample” and “spiked sample”.

3. RESULT AND DISCUSSION

3.1 Determination of the Pu, Am and Cm

Prior to the analysis of the spent nuclear fuel samples, an appropriate sample size was determined according to

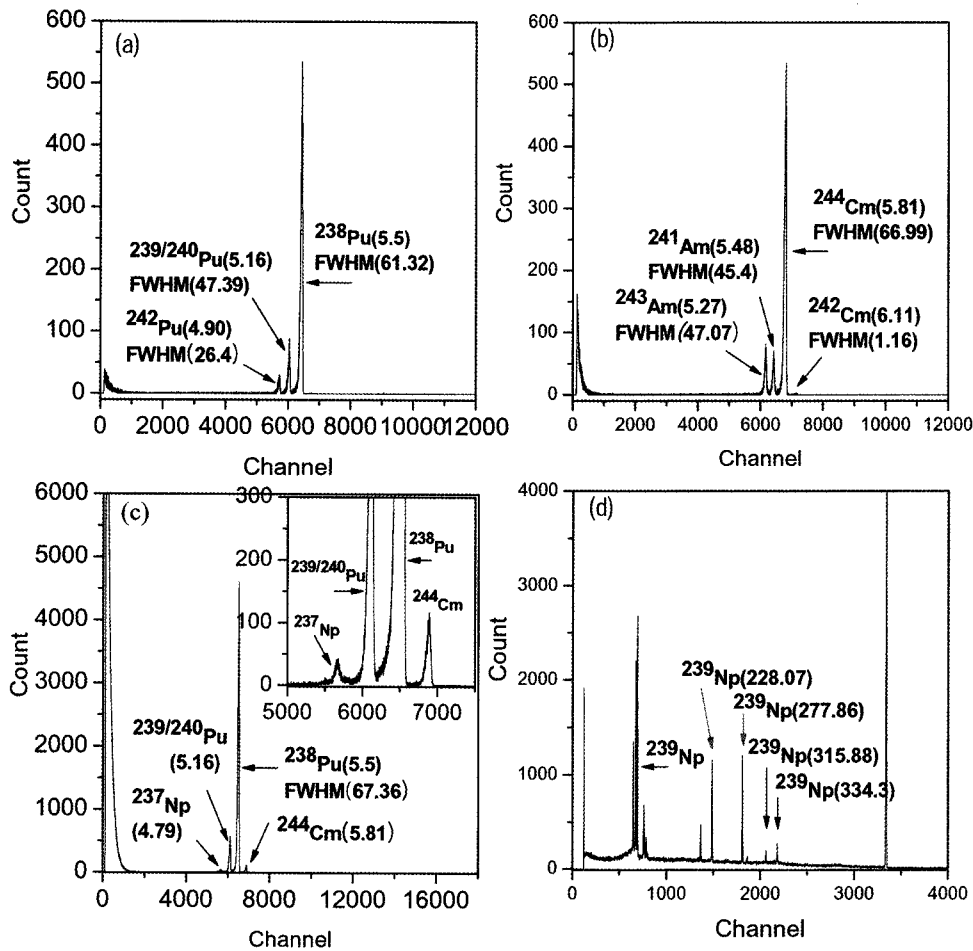


Fig. 2. Alpha Spectra and Gamma Spectrum of Transuranium Elements. (a) Pu Isotopes, (b) Am and Cm Isotopes, (c) ^{237}Np and (d) Gamma Spectrum of ^{239}Np , Sample : SF-1, Tracer; ^{242}Pu 2.258 Bq, ^{243}Am 15Bq, Counting Time; 5000 sec, Numbers in Parenthesis : MeV for Nuclides/KeV for FWHM (a, b, c) and KeV (d)

estimates [17, 18]. The alpha spectra (Fig. 2a) of the Pu isotopes in the spent fuel sample (SF-1) shows the clearly resolved three peaks of ^{238}Pu , $^{239+240}\text{Pu}$, and ^{242}Pu (yield tracer) at 5.5, 5.16, and 4.90 MeV, respectively. Good resolution (26-60 KeV full width at half maximum-FWHM) was obtained. The ^{239}Pu and ^{240}Pu content was obtained from analysis of the total activity of ^{239}Pu and ^{240}Pu . The total activity was divided into each nuclide by applying specific activities and isotope ratios measured by mass spectrometry after a collection of the Pu fraction at a separation step. Generally, for the measurement of ^{238}Pu in the uranium matrix samples, alpha spectrometry is recommended because of the ^{238}U - ^{238}Pu isobaric interference problematic in mass spectrometry. No ^{244}Cm peak in the plutonium fraction was observed (Fig. 2a). This means that the large ^{238}Pu peak (5.5 MeV) was not contaminated by the ^{241}Am peak (5.48 MeV) because the ^{244}Cm peak is generally much larger than the ^{241}Am peak or similar to that in spent fuels. The alpha spectra of the

Am and Cm isotopes was also measured (Fig. 2b). The ^{242}Cm , ^{244}Cm , ^{241}Am , and ^{243}Am peaks were also well resolved at 6.11, 5.81, 5.48, and 5.27 MeV, respectively. The ^{241}Am peak can be influenced by the ^{238}Pu peak (5.50 MeV) if the plutonium is contaminated. However, in this sample it was confirmed that the combined fraction of americium and curium was not contaminated by plutonium because of the absence of the 5.16 MeV $^{239+240}\text{Pu}$ peak.

The ^{243}Am was used for a recovery yield of the Am and Cm isotopes. A 1% error in the alpha spectrometry measurements is expected for these nuclides due to the original content of the ^{242}Pu and ^{243}Am tracers in the spent fuels samples [18]. The ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Am , ^{242}Cm , and ^{244}Cm measured contents in the six spent fuel samples were compared with results from the calculations (Table 2). The uncertainties associated with this work were evaluated using the standard solutions of ^{239}Pu , ^{241}Am , and ^{244}Cm in the uranium matrix, and they were less than 10 % at about 10 Bq for each nuclide [20].

Table 2. Comparison of Transuranium Elements between Measurements and Calculations in Spent fuel Samples

Sample	BU(GWD/MtU)	²³⁸ Pu(μg/mgU)			²³⁹ Pu(μg/mgU)			²⁴⁰ Pu(μg/mgU)		
		Meas.(m)	Cal.(c)	(m-c)100/m	Meas.(m)	Cal.(c)	(m-c)100/m	Meas.(m)	Cal.(c)	(m-c)100/m
SF-1	53.865	0.5598	0.3899	30.34	6.0002	6.2658	-4.43	3.5398	2.9738	15.99
SF-2	35.557	0.2742	0.1525	44.38	7.0482	5.8416	17.12	2.8106	2.1966	21.85
SF-3	52.525	0.5844	0.4166	28.72	6.4680	6.2582	3.24	3.5463	2.8854	18.64
SF-4	44.155	0.5179	0.2972	42.62	6.2328	6.0657	2.68	3.2792	2.5583	21.99
SF-5	52.688	0.5344	0.4190	21.60	5.7668	6.2618	-8.58	3.1454	2.8909	8.09
SF-6	51.288	0.5584	0.3984	28.66	6.6009	6.2315	5.60	3.5429	2.8426	19.77
Aver ± 1S				32.72 ± 8.90			2.61 ± 8.88			17.72 ± 5.21

Sample	BU(GWD/MtU)	²⁴¹ Am(μg/mgU)			²⁴² Cm(μg/mgU)			²⁴⁴ Cm(μg/mgU)		
		Meas.(m)	Cal.(c)	(m-c)100/m	Meas.(m)	Cal.(c)	(m-c)100/m	Meas.(m)	Cal.(c)	(m-c)100/m
SF-1	53.865	0.7444	0.5942	20.17	0.000006	0.000005	10.62	0.2977	0.1254	57.88
SF-2	35.557	0.6411	0.4219	34.18	0.000007	0.000004	45.37	0.0554	0.0150	72.86
SF-3	52.525	0.3605	0.2573	28.62	0.001887	0.001140	39.61	0.2661	0.1149	56.80
SF-4	44.155	-	-	-	0.001791	0.000791	55.82	0.2283	0.0601	73.69
SF-5	52.688	0.4276	0.2577	39.73	0.001833	0.001147	37.42	0.2699	0.1163	56.90
SF-6	51.288	0.5905	0.2542	56.96	0.001859	0.001086	41.60	0.2661	0.1047	60.64
Aver ± 1S				35.93 ± 13.8			38.41 ± 15.08			63.13 ± 7.99

Sample	BU(GWD/MtU)	²³⁷ Np(μg/mgU)		
		Meas.(m)	Cal.(c)	(m-c)100/m
SF-2	35.557	0.4510	0.5288	-17.27
SF-3	52.525	0.5267	0.8594	-63.16
SF-4	44.155	0.3487	0.6936	-98.91
SF-5	52.688	0.6709	0.8625	-28.56
Aver ± 1S				-51.97±36.88

done ahead of the Np elution during the separation procedure of Pu. These contaminated Pu peaks can also be reduced further by purification (repeating the full procedure). Note that this step might increase errors due to the resulting low recovery yield. The measured activities of ²³⁹Np and ²³⁷Np in the “sample” and “spiked sample” were used in equation (1) to obtain the amount of ²³⁷Np.

$$C_x = C_t \left(\frac{m_t}{m_x} \right) \left(\frac{M_x}{M_t} \right) \left\{ \left(\frac{R_t - R_m}{R_m - R_x} \right) \right\} \left(\frac{\sum_x R_i}{\sum_t R_i} \right) \quad (1)$$

Here, C_x and C_t denote the total concentration of Np in the “sample” solution and “spike” solution, m_t and m_x the weight of the “spike” solution and “sample” solution,

M_x and M_t the average atomic weight of Np in the “sample” solution and “spike” solution, R_t , R_m and R_x the isotope ratio of ²³⁷Np/²³⁹Np in the “spike” solution, “spiked sample” solution, and “sample” solution, $\sum_x R_i$ and $\sum_t R_i$ the sum of the isotope ratios of Np in the “sample” solution (²³⁷Np/²³⁹Np + ²³⁹Np/²³⁹Np) and “spike” solution (²³⁷Np/²³⁹Np + ²³⁹Np/²³⁹Np), respectively.

The uncertainty associated with the determination of ²³⁷Np in this work was presumed to be about 20%, which agrees with a previously calculated result [18].

3.3 Correlation with the ORIGEN-2 Code

In this study, the transuranic elements were determined

3.2 Determination of ²³⁷Np

For the determination of ²³⁷Np, ²³⁹Np was used as a spike by using the isotope dilution method because a spent nuclear fuel sample itself includes a considerable amount of ²³⁹Np. The measured ²³⁷Np alpha spectrum shows the Pu, Am, and Cm impurity peaks (Fig. 2), which did not interfere with the ²³⁷Np peak. The peaks in the gamma spectra of ²³⁹Np were larger in the “spiked sample” than in the non-spiked as expected (Fig. 2d). Insofar as the ²³⁹Np by gamma spectroscopy results were concerned, consideration of the intensities and branching ratios led us to select the 228.08 KeV and 277.86 KeV peaks as representative energies. In particular, the gamma activity of ²³⁹Np was measured as soon as possible after a separation from the samples due to its short half-life.

The high plutonium peaks in the Np fraction were reduced by allowing the columns to stand for about 2 hours to reduce the Pu(IV) or Pu(VI) to Pu(III). This was

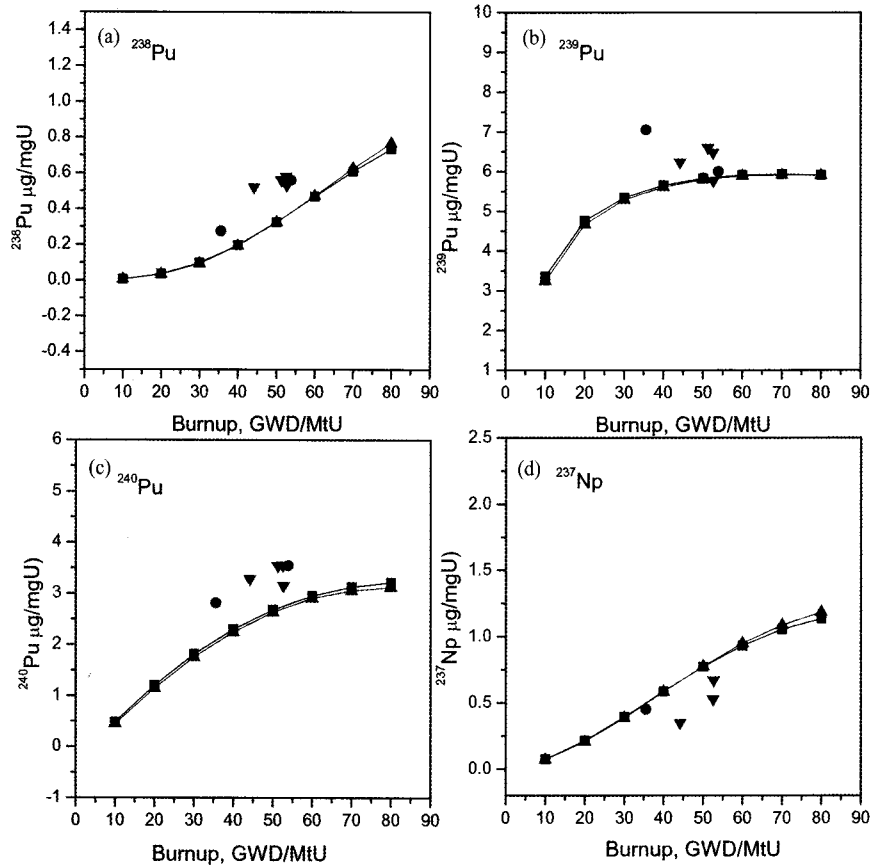


Fig. 3. Correlation of Pu isotopes (a, b, c) and ^{237}Np (d) between Measurements and Calculations in Spent Fuel Samples. ORIGEN-2 : \blacktriangle (2.1y)/ \blacksquare (6.3y) as a Cooling Time, Sample : SF-1 and SF-2 (\bullet , 6.3y)/SF-3 ~ SF-6 (\blacktriangledown , 2.1y)

by known analytical methods as mentioned above and were compared with calculations from the ORIGEN-2 code [17]. Burnups of sample specimens taken from different positions of the fuel (assembly) prior to the chemical measurements (Table 1) were determined by chemical methods using a ^{148}Nd monitor. Thus, we measured local burnups, not calculated average burnups. Nuclide contents measured from the spent fuel samples were compared with ORIGEN-2 calculations (Table 2), and correlations of the Pu and Np isotopes between the measured and calculated results as a function of burnup were examined (Fig. 3). The measured values of ^{238}Pu were in the range of 0.27-0.58 $\mu\text{g}/\text{mgU}$ and increased with burnup (Fig. 3a). The measurements were higher than the calculations by 32.7% on average. In the case of ^{239}Pu and ^{240}Pu (Figs. 3b and 3c), the differences between the measurements and calculations ranged from -8~17% resulting in a 3% difference on average and an 8-22% difference resulting in 18% on average, respectively. We performed two ORIGEN-2 calculations representing different sample cooling times (2.1 y and 6.3 y). The two curves showed nearly similar trends. The data for Pu measured in this work showed higher values (3-33%)

than the calculations according to the isotopes and these values showed similar trends comparing with those in other works. That is, the differences between measured values and calculated values (SAS2H code) for the samples from the Takaham-3 reactor [21] were about 13%, and the calculated values by ORIGEN-2 code for the PWR spent fuel samples showed about a 15% difference from the measured values.

The isotopic compositions of actinides were also computed by the CELL2 and PDQ codes[3]. This was done for the spent fuels of 44.34 GWD/MtU and 3.08% initial ^{235}U concentration for the implementation of burnup credit in spent fuel casks and for the U.S spent fuel inventory for fuel characterization [3]. The computed values were much biased from the experimental values such as -0.2, 3.7 and -8.1 as ratios of computed value/experimental data for ^{238}Pu , ^{239}Pu and ^{240}Pu , respectively. For the *Yankee Rowe* spent fuel samples [4], the SAS2H predictions agreed comparatively well with the experimental data as shown by the following ratios of calculation to measurement: -13.97~4.12% for ^{238}Pu , -9.35~9.92% for ^{239}Pu , and -8.2~3.13 % for ^{240}Pu . The reason for lower calculated values than the experimental measurements was explained due

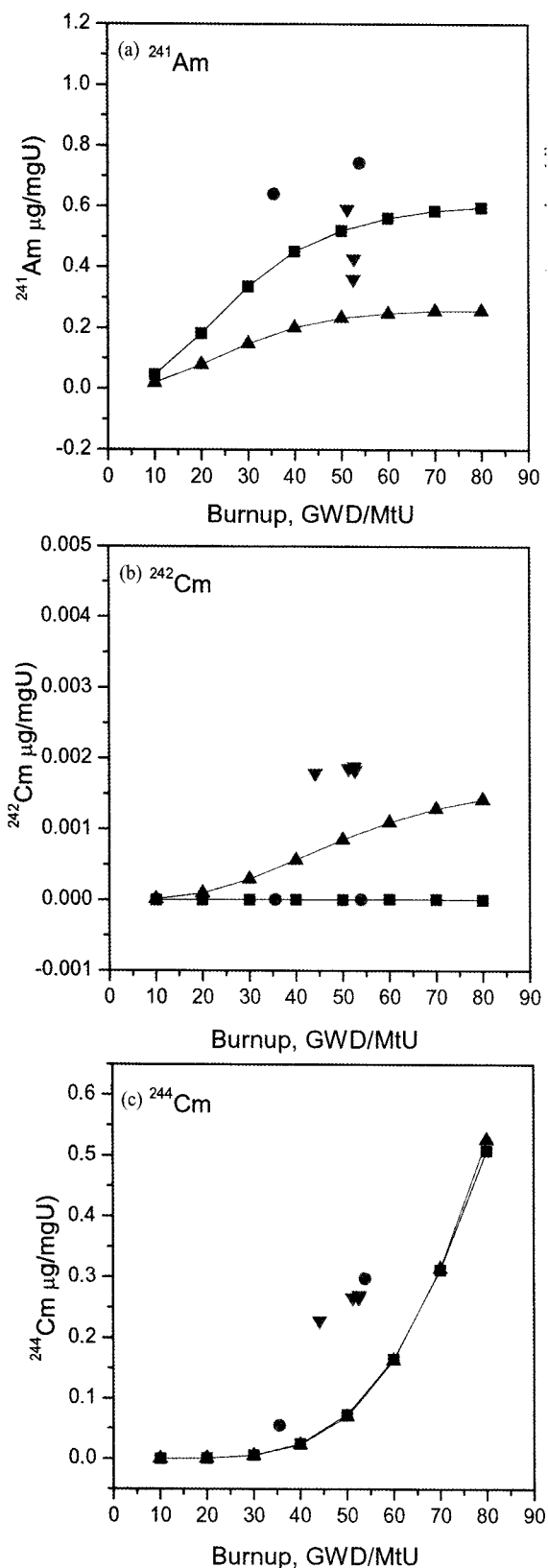


Fig. 4. Correlation of Am (a) and Cm Isotopes (b, c) between Measurements and Calculations in Spent Fuel Samples. ORIGEN-2 : \blacktriangle (2.1y) / \blacksquare (6.3y) as a Cooling Time, Sample : SF-1 and SF-2 (\bullet , 6.3y)/SF-3 ~ SF-6 (\blacktriangledown , 2.1y)

to the slightly harder (than what the code expected) neutron spectrum irradiating the fuel assemblies. Other SAS2H calculations for Pu isotopes (for burnup credit evaluation at Oak Ridge National Laboratory) exhibited good agreement (within 10%) with the experimental data for a variety of samples [5]. Correlations between the measurements and calculations for ^{237}Np was also investigated (Fig. 3d). The measured values were much lower than the calculations (-52%), and showed large scattered with a deviation of 36.9%. For the determination of ^{237}Np in the spent fuel samples, the differences between the measurements and calculations have been reported to be high to some extent according to the burnup. For the high-burnup PWR spent fuels, the measured and calculated (SAS2H) ^{237}Np was in good agreement (within 4%) [21]. However, for the low burnup fuels, agreement was worse (71.8%) [21]. Data regarding the ^{237}Np determined in MOX fuel irradiated up to 120 GWD/MtU in the Mark-II core of JOYO, Japan has been reported [23]. This data (for ^{237}Np at the core center of the MOX fuel) agreed well with the ORIGEN-2 calculation. However, larger uncertainties associated with the axial arrangement of a pin were observed [23].

In this study, some uncertainties associated with ^{237}Np measurements using ^{239}Np as a spike are expected because the activity ratio of ^{237}Np to ^{239}Np is excessively low in the spent fuel samples.

Correlations between the measurement and the calculation of Am and Cm are shown in Figure 4. Measured values for ^{241}Am exceeded the calculated values by about 36% on average (Fig. 4a). Since the presence of ^{241}Pu (in the spent fuel) causes a buildup of ^{241}Am over time, different trends as a function of burnup were observed for the two different cooling times of 2.1 and 6.3 y (Fig. 4a).

Figure 4b shows the correlations of ^{242}Cm between the measurements and the calculations as a function of the burnup. Different cooling times result in different correlations of ^{242}Cm (Fig. 4b). For long cooling times (6.3 y), most of the ^{242}Cm has decayed. Thus, the measurements and calculations are closely matched at the bottom level even though about a 38% difference between them occurred. The measured values of ^{244}Cm were larger than the calculations by about 63% with a fairly good precision (8%) (Fig. 4c).

4. CONCLUSION

Transuranic element content in high burnup (35.6~53.9 GWD/MtU) spent nuclear fuel samples was determined by alpha spectrometry. The measured nuclide contents were compared with ORIGEN-2 calculations.

The measured values of the plutonium isotopes were higher than the calculations by 3-33% on average. For the americium and curium isotopes, the measured values were higher than the calculations by about 35.9~63.1%

according to the nuclides, while for ^{237}Np , the measured values were lower than the calculations by about 52% on average with a large standard deviation (36%). Some differences between the experimental measurements and calculations were observed. At present, we do not have sufficient experimental data to explain all of the discrepancies with full confidence. However, some explanations might be attributed to the difference in neutron flux exposed to the fuel assemblies as compared to what the code calculated, problems with ORIGEN-2 input libraries, and even measurement uncertainty. Thus, it is important that we continuously build up on the existing experimental data.

In future information on the transuranic elements determined in this work will be used for spent fuel chemical characterization and code evaluation for high burnup spent fuels.

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