

## **Burnup Measurement of Spent $U_3Si/Al$ Fuel by Chemical Method Using Neodymium Isotope Monitors**

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### **Abstract**

The total burnup in the spent  $U_3Si/Al$  fuel samples from Hanaro reactor was determined by destructive methods using  $^{148}Nd$ , the sum of  $^{143}Nd$  and  $^{144}Nd$ , the sum of  $^{145}Nd$  and  $^{146}Nd$ , and the sum of total Nd isotopes ( $^{143}Nd$ ,  $^{144}Nd$ ,  $^{145}Nd$ ,  $^{146}Nd$ ,  $^{148}Nd$  and  $^{150}Nd$ ) monitors. The fractional( $^{235}U$ ) burnup in the spent fuel samples was also determined by U and Pu mass spectrometric method. The samples were dissolved in a mixture of 4 M HCl and 10 M  $HNO_3$  without any catalyst. The separation of U, Pu and Nd from the spiked and unspiked sample solutions was achieved by two sequential anion exchange separation methods. The isotope compositions of these elements, after their separation from the fuel samples were measured by mass spectrometry. The contents of the elements in the spent fuel samples were determined by isotope dilution mass spectrometric method(IDMS) using  $^{233}U$ ,  $^{242}Pu$  and  $^{150}Nd$  as spikes. The effective fission yield was calculated from the weighted fission yields averaged over the irradiation period. The difference between total burnup values determined by various Nd monitors were in the range of 1.8%.

**Key Words** : burnup measurement, Nd isotope monitor,  $U_3Si/Al$  fuel

### **1. Introduction**

One of the important parameters required for studies of a nuclear fuel is burnup, which is the number of fission per 100 heavy nuclide atoms(mass  $\geq 232$ ) initially present in the fuel. Various methods were developed to measure the burnup both by non-destructive and by destructive techniques. Destructive method, which is based

on the determination of specific nuclides by chemical analysis after appropriate separation of heavy elements and a monitor fission product, is widely used as a reference method to measure a burnup of spent fuel[1]. The use of the fission product  $^{148}Nd$  as a burnup monitor for nuclear fuel is generally accepted as a standard method since the issue of the ASTM method E 321-69[2]. The isotope  $^{148}Nd$  was selected mainly because its

fission yield is independent of the fissioning actinide, and because of its low thermal neutron capture cross section. But a serious drawback with  $^{148}\text{Nd}$  is its reported susceptibility to a  $^{147}\text{Nd}$  neutron capture effect. Maeck[3] postulated its thermal neutron capture cross section for  $^{147}\text{Nd}$  which would cause increasing amounts of  $^{148}\text{Nd}$  with increasing neutron flux. Therefore, the reported number of fissions exceeded the actual number of fissions because of excess  $^{148}\text{Nd}$  being produced from neutron capture on  $^{147}\text{Nd}$ . The ASTM method prescribes  $^{150}\text{Nd}$  as isotopic diluent, generally designated as "spike". The isotope  $^{142}\text{Nd}$  is used to monitor and correct for possible contamination of the fission product with natural Nd during the chemical separation. An approach is to use another monitor such as the sum of  $^{145}\text{Nd}$  and  $^{146}\text{Nd}$  because it appears invariant with neutron flux and fluence. While  $^{145}\text{Nd}$  has a significant capture cross-section, the  $^{145}\text{Nd}$  production cross-section from  $^{144}\text{Ce}$  and  $^{144}\text{Nd}$  is small and approximately equivalent to the burn-out cross-section on  $^{146}\text{Nd}$ .

The nuclear fuel currently used in the Hanaro reactor is a  $\text{U}_3\text{Si}/\text{Al}$  dispersion with an enrichment of 19.75 w/o[4]. The 19.75% enriched fuel meat consists of 61.4 wt.%  $\text{U}_3\text{Si}$ -38.6 wt.% Al made by a dispersion process of small particles of a high density uranium silicide compound in an aluminum matrix. The intermetallic compound  $\text{U}_3\text{Si}$  contains 3.96 wt.% silicon. The Hanaro fuel is made of pencils of an extruded  $\text{U}_3\text{Si}/\text{Al}$  dispersion with finned Al cladding and end plugs. Spent Hanaro fuel had high burnup values and the fuel could not be separated from its Al cladding, this eliminated the possibility of measuring the net weight of the fuel for the determination of the heavy element content. This fuel contained large amounts of fission products and Al from the fuel matrix both of which could be present in concentrations greater than that for the remaining unfissioned

heavy element content. The methods developed for the separation of these elements from the PWR fuel of  $\text{UO}_2$  type could not be used directly because of the disturbing influence of a large amount of Al in the dissolved  $\text{U}_3\text{Si}/\text{Al}$  fuel solutions. It was known that the chromatographic separation of the lanthanide fission products is most influenced by the presence of Al[6]. It was found that a satisfactory chromatographic separation of the Nd could be achieved only when Al is removed from the solution examined. The dispersed and metal alloy fuel can be dissolved in  $\text{HNO}_3$  using a mercury catalyst, applying a mixture of HCl and  $\text{HNO}_3$  without any catalyst or heating in 3-8 M NaOH[5-8].

The aim of this work is to determine the total burnup by using various Nd monitors on the same sample from spent  $\text{U}_3\text{Si}/\text{Al}$  fuels and to compare the results for the validity of the methods. In this paper experiments are described on the dissolution of the spent  $\text{U}_3\text{Si}/\text{Al}$  fuel sample for the burnup measurement, on the determination of the isotope compositions of U, Pu and fission product Nd after their separation by ion exchange separation techniques from the fuel samples and on the determination of the elements in the spent fuel by isotope dilution mass spectrometric method using  $^{233}\text{U}$ ,  $^{242}\text{Pu}$  and  $^{150}\text{Nd}$  as spikes.

## 2. Experimental

### 2.1. Chemicals and Instruments

All spikes( $^{233}\text{U}$ ,  $^{242}\text{Pu}$  and  $^{150}\text{Nd}$ ) certified were obtained from Oak Ridge National Laboratory(ORNL)(Table 1). The spike solutions were prepared by dissolving the oxides in  $\text{HNO}_3(1+1)$ -0.01 M HF. NBL CRM 129  $\text{U}_3\text{O}_8$  powder(New Brunswick Laboratory) and U standard solution(Spex Industries Inc.) were used as a reference standard material for U. Plutonium

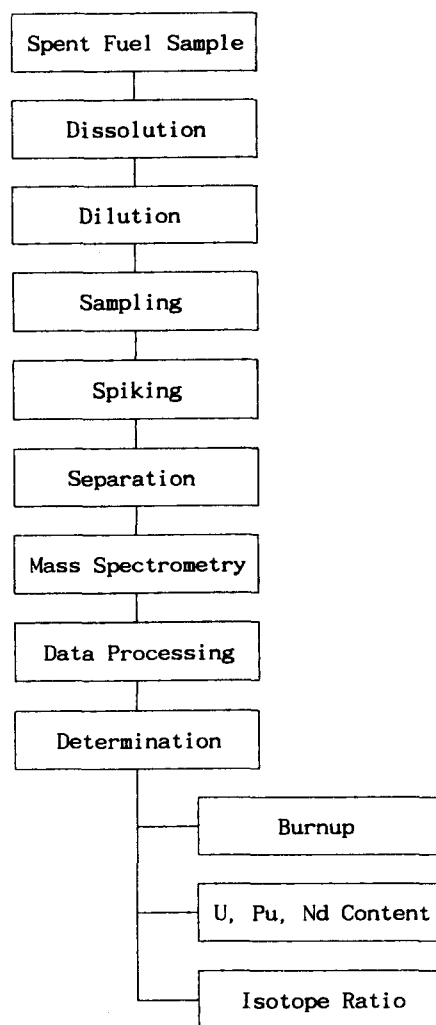
**Table 1. Isotopic Composition of Spikes from ORNL**

Spike	Isotope	Atom %
$^{233}U$	U-233	99.470
	U-234	0.166
	U-235	0.064
	U-236	0.015
	U-238	0.282
$^{242}Pu$	Pu-238	0.004
	Pu-239	0.005
	Pu-240	0.022
	Pu-241	0.035
	Pu-242	99.930
$^{150}Nd$	Nd-142	0.77
	Nd-143	0.39
	Nd-144	0.88
	Nd-145	0.34
	Nd-146	0.84
	Nd-148	0.66
	Nd-150	96.13

spike solution was standardized by coulometry. Neodymium standard solutions were obtained from AccuTrace and Spex Industries Inc.. The isotopic compositions of U, Pu and Nd separated from the spent  $U_3Si/Al$  fuel were determined using a mass spectrometer of the PINIGAN 262 type.

## 2.2. Spent Fuel Dissolution and Sample Preparation

Basic Processes in PIE analytical laboratory for the burnup determination are shown in Fig. 1. A spent fuel sample weighed exactly was placed in a 100 mL dissolution flask of dissolution apparatus with a separatory funnel[9,10]. The fuel sample was refluxed for more than 24 hours in 15 mL of 4 M HCl and 10 M  $HNO_3$  without catalyst. The fuel sample was placed in a dissolution flask and 5 mL of 4 M HCl was added dropwise, applying water

**Fig. 1. Basic Processes in PIE Analytical Laboratory**

cooling. After the Al had been dissolved (hydrogen bubbling ceased), 5 mL of 10 M  $HNO_3$  was added to the flask. This fuel solution was weighed and an aliquot was diluted with the  $HNO_3(1+1)$  to give a  $^{148}Nd$  concentration of about 200 ng per mL solution with the aid of ORIGEN[11] calculation for the estimation of nuclides content in the spent nuclear fuel. An aliquot of the diluted fuel solution was placed in a capped vial and transferred from the shielded facility into a glove box.

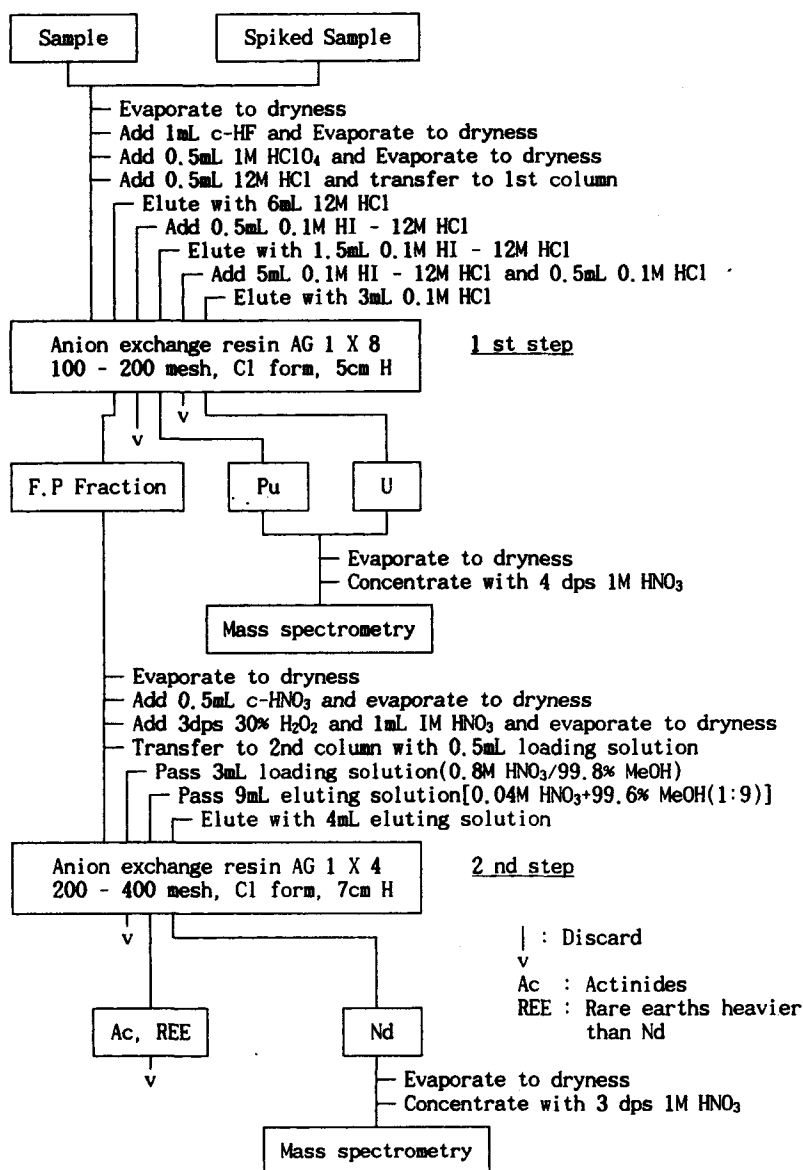


Fig. 2. Analytical Scheme for U, Pu and Nd Separation

### 2.3. Separation Procedure

Chemical separation was carried out for both the unspiked and the spiked sample solutions in the same experimental conditions in glove box without heavy shieldings. Two portions were

subjected to determine U, Pu and Nd isotopes in sample with and without spike addition followed by two sequential anion exchange separation procedures shown in Fig.2. The solutions were evaporated to dryness to assure complete U, Pu and Nd isotope exchange. Most of the light grey

residue from the fuel was volatilized by subsequent treatment with HF(possibly  $SiF_4$ ). The sample solution which contains less than 1 mg of U was redissolved with a half mL of 12 M HCl and then loaded on the first column. Neodymium and other fission products were passed through the column with 6 mL of 12 M HCl. Plutonium was eluted from the first column with 12 M HCl-0.1 M HI mixture, followed by U with 0.1 M HCl. The fission products fraction collected is chemically treated and is fed to the second column for pure Nd separation. After passing the sample solution through the column, the inner walls of the column were rinsed with 3 mL of loading solution[0.8 M  $HNO_3$ -90% MeOH], and All of Al and Si was passed immediately through the column. Neodymium was eluted with eluting solution[0.04 M  $HNO_3$  : 99.8% MeOH(1:9)]. The effluent from 8 to 11 mL was taken as the Nd fraction. Isotopic compositions of U, Pu and Nd in each fractions isolated and then concentrated were measured by the thermoionization mass spectrometer(TIMs). Each sample solutions of U, Pu and Nd in the range of  $\mu g$  to ng were loaded on a triple rhenium filament and then measured.

## 2.4. Correction of Nd Isotopes

The mass discrimination bias factor(B) for all Nd isotopes measured was calculated as follows[1] :

$$B = (1/C) [(R_{ab}/R_m) - 1] \quad (1)$$

$$R_{cab} = R_{ab} (1 + CB)$$

where, B : mass discrimination bias factor,

C :  $\Delta mass/mass$ ,

$R_m$  : known value of the measured atom ratio,

$R_{ab}$  : average measured atom ratio of isotope a to isotope b,

$R_{cab}$  : corrected average atom ratio of

isotope a to isotope b.

The contribution of natural Nd for all Nd isotopes measured was corrected by the equation(2)[1,12]:

$$R = \frac{A_{142} - A_{148}({}^{142}\text{Nd}/{}^{148}\text{Nd})}{A_{142}({}^{150}\text{Nd}/{}^{148}\text{Nd}) - A_{150}({}^{142}\text{Nd}/{}^{148}\text{Nd})} \quad (2)$$

where, R is the corrected  ${}^{148}\text{Nd}/{}^{150}\text{Nd}$  ratio, and A is the natural abundance of the Nd isotopes.

## 2.5. Calculation of ${}^{235}\text{U}$ Burnup

Fractional( ${}^{235}\text{U}$ ) burnup( $F_5$ ) value in atom % fission was calculated by the mass spectrometric method according to the equations(3)and(4)[13,14] :

$$\text{Atom \% fission}(F_5) = N^0 8 [(R^0 5/8 - R5/8) - (R6/8 - R^0 6/8)] \quad (3)$$

$$\text{Atom \% fission}(F_5) = N^0 8 [(R^0 5/8 - R5/8) / (1 + \alpha_5)] \quad (4)$$

where,  $N^0 8$  : atom % abundance of  ${}^{238}\text{U}$  in the pre-irradiated fuel sample

$R^0 5/8$  : atom ratio of  ${}^{235}\text{U}$  to  ${}^{238}\text{U}$  in the pre-irradiated fuel sample

$R^0 6/8$  : atom ratio of  ${}^{236}\text{U}$  to  ${}^{238}\text{U}$  in the pre-irradiated fuel sample

$R5/8$  : atom ratio of  ${}^{235}\text{U}$  to  ${}^{238}\text{U}$  in the post-irradiated fuel sample

$R6/8$  : atom ratio of  ${}^{236}\text{U}$  to  ${}^{238}\text{U}$  in the post- irradiated fuel sample

$\alpha_5$  : capture-to-fission cross section ratio for  ${}^{235}\text{U}$

## 2.6. Calculation of Effective Fission Yield and Total Burnup

Total burnup( $F_t$ ) value in atom % fission was calculated using different Nd isotope monitors according to the equation(5)[1] :

$$\text{Atom \% fission}(Ft) = \frac{N/Y}{N/Y + N(U) + N(Pu)} \times 100 \quad (5)$$

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

Y value is provided by the weighted average fission yield calculated according to the equations (6) and (7)[15] :

$$Y = f \cdot Y_{\text{ajw}}(\text{thermal}) + (1-f) \cdot Y_{\text{ajw}}(\text{fast}) \quad (6)$$

where, f : fraction of thermal fissions

$Y_{\text{ajw}}(\text{thermal})$  : weighted average thermal fission yield for isotope x

$Y_{\text{ajw}}(\text{fast})$  : weighted average fast fission yield for isotope x

$$Y_{\text{ajw}} = 1/2 \left[ \left( \sum N_i \alpha \gamma / \sum N_i \alpha \right)_B + \sum N_i \alpha \gamma / \sum N_i \alpha \right]_E \quad (7)$$

where,  $Y_{\text{ajw}}$  : weighted average fission yield for isotope x

$\gamma$  : fission yield for fissionable isotope i

$N_i$  : concentration of fissionable isotope i

$\alpha$  : fission cross section for fissionable isotope i

B : at the beginning of the irradiation

E : at the end of the irradiation

### 3. Results and Discussion

#### 3.1. Dissolution of Spent $U_3Si/Al$ Fuel

In our previous work[9,10], we studied the dissolution of nuclear fuel based on unirradiated  $U_3Si/Al$  in 6 M  $HNO_3$ , 6 M  $HNO_3$  using mercury

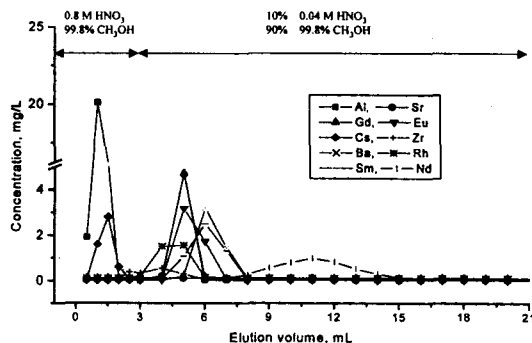
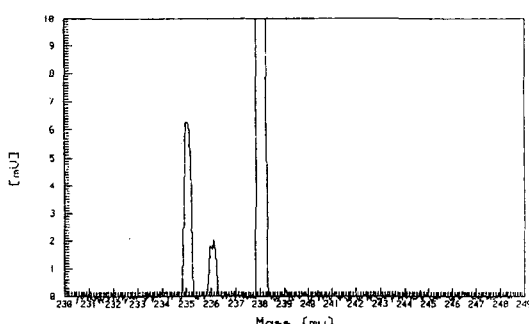


Fig. 3. Elution Curve of Nd for a Simulated  $U_3Si/Al$  Fuel

catalyst and applying a mixture of 4 M HCl and 10 M  $HNO_3$ . As a result, the latter was applied in this experiment for the dissolution of real spent fuel from the evaluation for the interference by the presence of mercury ions in dissolved solution, the rate of dissolution and the release of  $NO_x$  gas, and the amount of a residue remained in the solution. The dissolution of the  $U_3Si/Al$  dispersion fuel was slow and therefore continued for 24 hours for complete dissolution of all the Al. The fuel solution contained a small amount of residue of light grey. It is believed that the major components of this residue are noble element fission products[7].

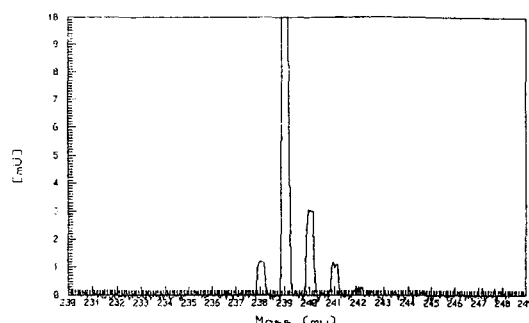
#### 3.2. Separation Procedure

In our previous work[9,10], a series of the preliminary experiments on the separation of U, Si, Al and some fission products were performed using the prepared stock solution of the  $U_3Si/Al$ . To the solution dissolved of unirradiated  $U_3Si/Al$ , suitable simulated fission products were added and their behaviour was studied in separation systems. As a result we found that a satisfactory chromatographic separation of these elements could be achieved. Fig. 3 shows the elution curves obtained by using the second anion exchange column with sample of unirradiated fuel solution



**Fig. 5. Mass Spectrum of U Separated from a Spent  $U_3Si/Al$  Fuel**

and the Nd was well separated from the other fission product elements in the sample. In the same manner the Nd in real fuel sample was purely separated from the other rare earth elements, Am and Cm by two sequential anion exchange separation procedures shown in Fig. 2. In this experiment the separation from Ce and Sm, which disturbed determination of Nd by IDMS was complete. Prior to load on the first column, treatment with small amount of 1 M  $HClO_4$  was used to assure all the Pu in  $Pu(VI)$  state because  $Pu(VI)$  is more strongly adsorbed on an anion exchange resin as negatively charged chloride complex form. Elution of the Pu with 12 M  $HCl$ -0.1 M  $HI$  mixture and the U with 0.1 M  $HCl$  were performed satisfactorily, and no further purification was needed. The Am and Np did not appear in the Pu and U portions and the complete separation was identified from the mass spectra of Fig. 4 and 5. The U, Pu and Nd portions isolated were dried and then transformed into the nitrate form to analyze by mass spectrometry. Major advantages in this chromatographic separation were the use of single eluent agent, needless of additional purification steps in other separation procedure, expecting the possibility of sequential separation, and room temperature operation.



**Fig. 4. Mass Spectrum of Pu Separated from a Spent  $U_3Si/Al$  Fuel**

### 3.3. Determination of Isotopic Composition

The U, Pu and Nd fractions collected from the spiked ( $^{233}U$ ,  $^{242}Pu$  and  $^{150}Nd$ ) and unspiked fuel samples were prepared for mass spectrometric determination. In the calibration of the mass spectrometer for the isotope analysis, the correction of mass discrimination is an important factor to obtain accurate and consistent results. The mass discrimination bias factor,  $B$ , is constant for a given method of scanning and detection on a given detector. The mass discrimination bias can be readily measured on natural Nd according to the equation (1)[1]. In this work, all measured average ratios of Nd were corrected for mass discrimination to achieve high accuracy on burnup measurement.

The contributions to the isotopic composition of fission product Nd are shown in Fig. 6. A pure Nd fraction was obtained from the samples containing less than 1  $\mu g$  of Nd by the separation procedure shown in Fig. 2. This is very important because the isobaric pair,  $^{150}Nd + ^{150}Sm$ , interferes directly to the isotope dilution mass spectrometry, and also  $^{142}Ce$  disturbs the  $^{142}Nd$  determination that is essential to correct the contribution of contamination with natural Nd, since  $^{142}Nd$  is not formed by fission. The presence of other rare

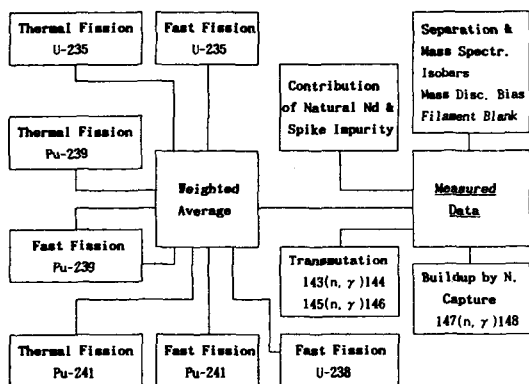


Fig. 6. Contribution to the isotopic composition of fission product Nd

earth isobars,  $^{142}\text{Ce}$  and  $^{148}\text{Sm}$ , in the Nd fraction can introduce a bias in the  $^{148}\text{Nd}$  determination because these isobars can affect the measurement of the  $^{148}\text{Nd}/^{142}\text{Nd}$  ratio. Both  $^{142}\text{Ce}$  and  $^{148}\text{Sm}$  are naturally occurring isotopes and also are formed in the fission processes. The contribution of these isobars to the Nd fraction can be identified by monitoring the mass 140, 147 and 149 peak[16].

Table 2 shows the isotopic compositions of U, Pu and Nd in a  $\text{U}_3\text{Si}/\text{Al}$  spent fuel samples measured by the thermoionization mass spectrometer. In case of Nd the corrections for mass discrimination and contribution of natural contamination were made for all isotopes. After the mass spectrometric measurement and the correction for their isotope compositions of U, Pu and Nd portions isolated from a spiked and a unspiked sample solution, the concentrations of U, Pu and Nd in sample solution were determined by the isotope dilution method (Table 3). A major advantage in IDMS is that quantitative recovery of the elements concerned is not required.

### 3.4. Determination of $^{235}\text{U}$ Fractional Burnup

Fractional( $^{235}\text{U}$ ) burnup,  $F_5$  can be calculated from equation (3) assuming no loss of  $^{238}\text{U}$  or  $^{236}\text{U}$

Table 2. Isotopic Compositions of U, Pu and Nd Separated from a  $\text{U}_3\text{Si}/\text{Al}$  Spent Fuel

U		Pu		Nd	
Isotope	Atom %	Isotope	Atom %	Isotope	Atom %
U-234	0.1194	Pu-238	2.8741	Nd-143	22.2856
U-235	8.1105	Pu-239	71.1231	Nd-144	32.0714
U-236	2.4883	Pu-240	17.9439	Nd-145	17.8632
U-238	89.2819	Pu-241	6.5650	Nd-146	15.8403
		Pu-242	1.4938	Nd-148	8.6969
				Nd-150	3.2426

Table 3. Determination of U, Pu, Nd and their Isotopes in a Spent  $\text{U}_3\text{Si}/\text{Al}$  Fuel Sample by Isotope Dilution Mass Spectrometry

U		Pu		Nd	
Isotope	Measured ( $\mu\text{g}$ )	Isotope	Measured ( $\mu\text{g}$ )	Isotope	Measured ( $\mu\text{g}$ )
U-234	0.1481	Pu-238	0.0172	Nd-143	0.4279
U-235	10.1069	Pu-239	0.4269	Nd-144	0.6274
U-236	3.1140	Pu-240	0.1082	Nd-145	0.3517
U-238	112.6822	Pu-241	0.0397	Nd-146	0.3035
		Pu-242	0.0091	Nd-148	0.1676
				Nd-150	0.0649
Total	126.0512		0.6011		1.9430

during irradiation. The equation (3) is best applied at high exposure, where more than 10% of the  $^{235}\text{U}$  is consumed. On the other hand, this equation is seriously affected by natural U contamination in the spent fuel sample, and for this reason, every precaution must be taken to prevent contamination from U in the chemistry and mass spectrometry operations. The samples for mass spectrometry should be larger than  $50 \mu\text{g}$  where it is possible to minimize the effect of such contamination. The principal merit in equation (3) lies in its independence of the reactor variable,  $\alpha$  [effective ratio of  $^{235}\text{U}(n, \gamma)$  capture-to-fission cross sections]. The  $F_5$  can also be calculated from equation (4) assuming no loss of  $^{238}\text{U}$  during irradiation. The main advantage of this equation is



**Table 4. Determination of Fractional Burnup( $F_5$ ), Depletion( $D_5$ ) and Alpha Value( $\alpha_5$ ) for  $^{235}U$** 

Sample	$F_5$ (Atom % Fission)		$D_5$	$\alpha_5$
	A	B		
L1-M	10.4497	10.6095	1.6834	0.1952
L1-L	8.3666	8.4997	2.1226	0.2070
L1-H	5.1167	5.1987	3.5130	0.2274

A : Values calculated according to equation (3)

B : Values calculated according to equation (4)

its independence from  $^{236}U$  and hence it is more reliable for the burnup determination of fuels with high pre-irradiated  $^{236}U$  content. The equation (4) requires a knowledge of  $\alpha_5$  and is best applied where more than 10% of the  $^{235}U$  is consumed. Serious errors can be introduced by natural U contamination in the spent fuel sample. In this work,  $\alpha_5$  was calculated from equation (8) with atom ratios and number of atoms determined by IDMS[17].

$$\alpha_5 = (R - R^0)A_5 / (P/Y) = \text{captures/fissions} \quad (8)$$

where, R : atom ratio of  $^{236}U / ^{235}U$  in the post-irradiated fuel sample

$R_0$  : atom ratio of  $^{236}U / ^{235}U$  in the pre-irradiated fuel sample

$A_5$  : number of  $^{235}U$  atoms in the post-irradiated fuel sample

P : number of atoms of fission product monitor( $^{148}Nd$ )

Y : fission yield for P

Table 4 shows  $F_5$  values calculated according to equations (3) and (4) together with  $\alpha_5$  and  $D_5$  values. The depletion of  $^{235}U$ ( $D_5$ , the difference between the initial and final  $^{235}U$  content) is expressed in  $W_5^0/W_5^0 - W_5$ . The  $F_5$  values from two equations showed good agreement within 1.6% difference.

### 3.5. Determination of Effective Fission Yield and Total Burnup

Walker[18] reported that the variance in the  $^{148}Nd$  isotopic abundance is greater than that of  $^{145}Nd + ^{146}Nd$  and even  $^{150}Nd$ , each of which are little affected by the neutron capture processes. That is, the calculated number of fissions exceeded the actual number of fissions because of excess  $^{148}Nd$  being produced from capture on  $^{147}Nd$ . The magnitude of this error is a function of the flux and fluence. This is especially significant for constant prolonged high flux irradiations, but a significant fraction of reactor down time will reduce the magnitude of the capture effect[19]. Another approach is to use another monitor such as the sum of  $^{145}Nd$  and  $^{146}Nd$  because it appears invariant with neutron flux and fluence. In thermal reactor spectra, considerable burnout of  $^{143}Nd$  and  $^{145}Nd$  occurs to  $^{144}Nd$  and  $^{146}Nd$ , respectively. Therefore, their fission yields and their abundances in the fission product Nd have to be added. In addition, if short-cooled fuel is examined, a substantial fraction of the mass 144 is still present as  $^{144}Ce$ , and the abundance of  $^{144}Nd$  should be corrected accordingly.

In this work, burnup values(in atom % fission) of  $U_3Si/Al$  fuel samples by various Nd isotope monitors were calculated according to a procedure in reference[1]. The concentration of U, Pu and Nd isotopes were simultaneously determined by IDMS. Neodymium-148, the sum of  $^{143}Nd$  and  $^{144}Nd$ , the sum of  $^{145}Nd$  and  $^{146}Nd$ , and the sum of total Nd isotopes( $^{143}Nd$ ,  $^{144}Nd$ ,  $^{145}Nd$ ,  $^{146}Nd$ ,  $^{148}Nd$  and  $^{150}Nd$ ) were used as a fission monitor. The successful application of this technique requires accurate measurements of the fission product monitor and heavy atoms and an accurate value for the effective fission yield. The effective fission yield was calculated from the weighted fission yields averaged over the irradiation period by the

**Table 5. Estimated Effective Fission Yield for Spent U<sub>3</sub>Si/Al Fuel Samples**

Fission Product	Effective Fission Yield		
	L1-M	L1-L	Li-H
Nd-143	5.8386	5.8537	5.8673
Nd-144	5.3751	5.3916	5.4022
Nd-145	3.8562	3.8647	3.8727
Nd-146	2.9581	2.9627	2.9671
Nd-148	1.6771	1.6769	1.6768
Nd-150	0.6741	0.6704	0.6672

**Table 6. Total Burnup in Atom % Fission Calculated for Different Nd Isotope Monitors**

Sample	Isotope	Atom % Fission	Difference
L1-M	Nd-148	11.2577	1.000
	Nd-(145+146)	11.1167	0.987
	Nd-(143+144)	11.0534	0.982
	Nd-Total	11.0603	0.982
L1-L	Nd-148	9.0301	1.000
	Nd-(145+146)	8.9822	0.995
	Nd-(143+144)	8.9269	0.989
	Nd-Total	8.9093	0.987
L1-H	Nd-148	5.5145	1.000
	Nd-(145+146)	5.4967	0.997
	Nd-(143+144)	5.4666	0.991
	Nd-Total	5.4629	0.991

Total : 143+144+145+146+148+150

equations (6) and (7). The concentration of fissionable isotopes needed for the calculation was determined by IDMS, and the nuclear data were obtained from references[4,20,21]. Table 5 gives the effective fission yields for <sup>143</sup>Nd, <sup>144</sup>Nd, <sup>145</sup>Nd, <sup>146</sup>Nd, <sup>148</sup>Nd and <sup>150</sup>Nd isotopes calculated from the spent U<sub>3</sub>Si/Al fuel samples. Table 6 gives the total burnup(atom % fission) measured by various monitors, <sup>143</sup>Nd + <sup>144</sup>Nd, <sup>145</sup>Nd + <sup>146</sup>Nd, <sup>148</sup>Nd, and the sum of the total Nd isotopes. From the consideration of the irradiation characteristics and cooling time for these fuels, the abundance of <sup>144</sup>Nd was calculated without correction for <sup>144</sup>Ce.

The number of fissions by <sup>148</sup>Nd monitor was calculated without correction for excess <sup>148</sup>Nd produced from capture on <sup>147</sup>Nd. The data in Table 6 were in agreement within 1.8% for the same fuel sample. The use of all the Nd isotopes in the determination of burnup for nuclear fuel has the advantage of confirming the value obtained for <sup>148</sup>Nd. No additional separation work or mass spectrometric analysis is needed. The agreement of the number of fissions calculated from isotopes sensitive to fuel composition confirms the fissile isotope content.

For the most accurate burnup analysis, the requester should provide to the measurer an estimate of the neutron flux and irradiation history with respect to reactor up and down time, from which the appropriate correction factor can be calculated. The reactor cycling time could be important if the downs were frequent and the durations significant compared to the 11 d half life of <sup>147</sup>Nd. The fraction of the down time will reduce the magnitude of the correction.

#### 4. Conclusions

The use of all the Nd isotopes in the determination of burnup for nuclear fuel needs no increase in the work for chemical separation or mass spectrometric analysis. The advantage of considering the whole set of Nd isotopes lies primarily in the confirmation of the value obtained for <sup>148</sup>Nd. The agreement of the number of fissions calculated from isotopes sensitive to fuel composition(<sup>143</sup>Nd, <sup>144</sup>Nd, <sup>145</sup>Nd, <sup>150</sup>Nd) and values from isotopes which are practically independent from fuel composition(<sup>146</sup>Nd, <sup>148</sup>Nd) confirms the fissile isotope content of the fuel. The Nd isotope pattern provides information on the real irradiation characteristics which is necessary for evaluating the fuel performance in the reactor.

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