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Analysis of the Irradiated Nuclear Fuel Using the Heavy Atom and Neodynium Isotope Correlations with Burnup

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Abstract

The correlation of isotope composition of uranium, plutonium and neodymium with the burnup in PWR uranium dioxide fuel has been investigated experimentally. The total and fractional(^{235}U) burnup were determined by Nd-148 and, U and Pu mass spectrometric method respectively. 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 irradiated fuel were determined by isotope dilution mass spectrometric method using ^{233}U, ^{242}Pu and ^{150}Nd as spikes. The content of plutonium in the irradiated fuel was expressed by the correlation with uranium isotopes. The correlations between isotope compositions themselves and the total and fractional burnup were compared with those calculated from ORIGEN2 code.

1. Introduction

In order to check the consistency of post-irradiation analysis results, correlations between parameters of irradiated nuclear fuels such as concentration of heavy elements and fission products, ratios of their isotopes and burnup were established[1-23]. These correlations can be used to identify reactor fuels and to estimate the burnup and Pu production. Some of these correlations may also be useful for safeguards purposes. The isotope correlations can be divided into three groups, i. e. , correlation based on heavy isotopes and elements, stable fission products, and radioactive fission products. Among the elements used in these studies Kr, Xe, Cs, Nd, Eu, Ru, Zr, U and Pu should be mentioned[1,2].

The parameters generally used for the correlation studies are as follows ; total burnup, ^{235}U burnup, ^{235}U depletion, ^{239}Pu buildup, Pu/U ratio, U/U_0(U_0 : initial uranium content) ratio and some fission product ratios in the irradiated fuel such as $^{134}Cs/^{137}Cs$, $^{134}Cs/^{133}Cs$, $^{154}Eu/^{137}Cs$, $^{154}Eu/^{155}Eu$, $^{84}Kr/^{83}Kr$, $^{86}Kr/^{83}Kr$, $^{84}Kr/^{86}Kr$, $^{132}Xe/^{131}Xe$, $^{134}Xe/^{131}Xe$, $^{133}Xe/^{134}Xe$, $^{136}Xe/^{134}Xe$, $^{144}Nd/^{143}Nd$, $^{143}Nd/^{145+146}Nd$, $^{146}Nd/^{145}Nd$, $^{148}Nd/^{145}Nd$, $^{146}Nd/^{148}Nd$, $^{148}Nd/^{145+146}Nd$, $^{150}Nd/^{148}Nd$, $^{101}Ru/^{104}Ru$, $^{93}Zr/^{96}Zr$ and $^{91}Zr/^{96}Zr$[2-15]. The correlations between the different heavy element isotopes, total and ^{235}U burnup, ^{235}U and ^{239}Pu depletion, and Pu/U and U/U_0 mass ratio have also been reported [1,3-5,10,12,14-18]. The dependences of U isotope composition are characterized by a decrease of ^{235}U and by an increase of ^{236}U and ^{238}U with increasing burnup. The dependences of Pu isotope composition on burnup are characterized by a decrease of ^{239}Pu and an increase of ^{240}Pu, ^{241}Pu and ^{242}Pu. In this work, the dependences of U, Pu and

Nd isotope composition on burnup values, and correlations between U and Pu and their isotopes for the irradiated PWR uranium dioxide fuels from Kori-1 reactor were characterized experimentally.

2. Experimental

2.1. Chemicals and Mass Spectrometric Measurements

Basic Processes in PIE Analytical Laboratory for the burnup determination and isotope correlation analysis are shown in Fig. 1. The irradiated fuel samples were dissolved in HNO_3(1+1) in a lead shielded line. From the dissolved solution, an aliquot was taken and diluted for the measurement of U, Pu and Nd isotope composition for burnup determination. The separation of each element from fuel solution was carried out in a glove box using the separation procedure in our previous work[23]. The concentrations of U, Pu and Nd in the fuel solution were determined by a mass spectrometric isotope dilution method using ^{233}U, ^{242}Pu and ^{150}Nd as spikes certified

Table 1. Isotopic Composition of Spikes from ORNL

Spike	Isotopic Composition (Atom %)	
^{233}U	U-233	99.540
	U-234	0.184
	U-235	0.062
	U-236	0.013
	U-238	0.203
^{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

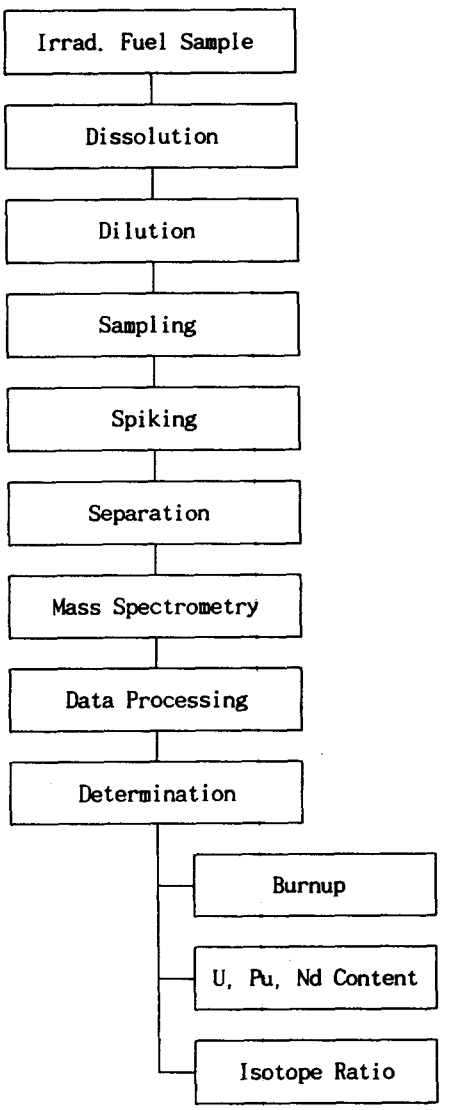

Fig. 1. Basic Processes in PIE Analytical Laboratory

from Oak Ridge National Laboratory(Table 1). NBS 950b U_3O_8 powder(National Institute of Standards and Technology, U.S.A.) was used as a reference standard material for U. Pu standard solution was supplied from Amersham International Ltd. (Amersham, UK) and Nd standard solution was obtained from Spex Industries Inc. (U.S.A.). Mass spectrometric measurements were performed by a TSN 206 SA(CAMECA, France) mass spectrometer equipped

with 90° sector magnetic analyzer, with a thermoionization source, an electron multiplier detector, and with Hewlett Packard HP 9825 data acquisition and processing system. U, Pu and Nd in the range of μg to ng were loaded on a Re filament, and then measured.

2.2. Calculation of Burnup

Total burnup(F_t) value in atom % fission was calculated by the Nd-148 method[12,23,24] according to the equation(1) :

$$\text{Atom \% fission}(F_t) = \frac{N/Y}{N/Y + N(U) + N(Pu)} \times 100 \quad (1)$$

where, N : the number of the monitor ^{148}Nd atoms in irradiated fuel solution,

Y : the effective fission yield of ^{148}Nd from the fissile elements,

N(U), N(Pu) : the number of U and Pu atoms in irradiated fuel solution, respectively.

Fractional(^{235}U) burnup(F_5) value in atom % fission was calculated by the mass spectrometric method[25] according to the equation(2) :

$$\text{Atom \% fission}(F_5) = N^o8[(R^o5/8 - R5/8) - (R6/8 - R^o6/8)] \quad (2)$$

where, N^o8 : heavy element atom % ^{238}U in the preirradiated fuel

$R^o5/8$: atom ratio of ^{235}U to ^{238}U in the preirradiated fuel

$R^o6/8$: atom ratio of ^{236}U to ^{238}U in the preirradiated fuel

R5/8 : atom ratio of ^{235}U to ^{238}U in the final irradiated sample

R6/8 : atom ratio of ^{236}U to ^{238}U in the final irradiated sample

2.3. Correction of Neodymium Isotopes

The contribution of natural Nd for all Nd isotopes measured was corrected by the equation(3)[14] :

$$R = \frac{A_{142} - A_{148}(^{142}Nd/^{148}Nd)}{A_{142}(^{150}Nd/^{148}Nd) - A_{150}(^{142}Nd/^{148}Nd)} \quad (3)$$

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

The isotope correlations between the heavy elements and Nd isotopes of interest were plotted against the burnup values measured experimentally and also the values calculated from the ORIGEN2 code.

3. Results and Discussion

3.1. Determination of Isotopic Composition

In the calibration of the mass spectrometer for the analysis of U, Pu and Nd, 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, and can be calculated as follows :

$$B = (1/C)\,[(R_{ab}/R_m) - 1] \quad (4)$$

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

where, B : mass discrimination bias factor,

C : △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 factor B for the natural Nd isotopes from the measured atom ratio of $^{148}Nd/^{150}Nd$ and $^{142}Nd/^{150}Nd$ were found to be 7×10^{-5} and 8×10^{-4}, respectively. These values were in the range of standard deviation in mass spectrometry. Therefore, it was not necessary to conduct the correction of mass discrimination bias for the measured atom ratio.

A pure Nd fraction was obtained from the samples

Table 2. Contribution of Other Isobars for Neodynium Isotopes

Mass	Isobars				
142	Nd*	Ce*	Ce+		
143	Nd*	Nd+			
144	Nd*	Nd+	Pr+	Sm*	Ce+
145	Nd*	Nd+			
146	Nd*	Nd+			
147	Sm*	Sm+	Pm+		
148	Nd*	Nd+	Sm*	Sm+	
149	Sm*	Sm+			
150	Nd*	Nd+	Sm*	Sm+	

*: natural

+ : fission product

containing less than 1 μg of Nd by the separation procedure[23]. 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 earth isobars, ^{142}Ce and ^{148}Sm, in the Nd fraction can introduce a bias in the ^{148}Nd determination because these isobars can affect the measurement of the ^{148}Nd/^{142}Nd ratio. Both ^{142}Ce and ^{148}Sm are naturally occurring isotopes and also are formed in the fission processes(Table 2). The contribution of these isobars to the Nd fraction can be identified by monitoring the mass 140, 147 and 149 peak. The mass 140 peak is just due to Ce content in the isolated Nd fraction because no other isotopes of mass 140 have a sufficiently long half-life to be present in irradiated fuel after decay of a few months. The natural ^{142}Ce/^{140}Ce ratio is 0.13 while the ratio of the fission yields for the mass 142 and mass 140 chains is 0.92. Thus, in case that the 142/140 ratio is large, the contribution of ^{142}Ce in the mass 142 peak can be ignored and therefore the bias by isobaric contamination can be negligible. From the mass spectrometric result of a Nd fraction separated from irradiated fuel, the 142/140 ratio was calculated to be 37.3.

Table 3. Determination of Uranium, Plutonium, Neodymium and their Isotopes in a PWR Fuel Sample by Isotope Dilution Mass Spectrometry

Isotope	Measured (μg)	Calculated* (μg)
^{234}U	0.0928	0.0028
^{235}U	4.7402	4.3011
^{236}U	2.0366	2.3275
^{238}U	438.9564	504.8677
Total	445.8260	511.4991
^{238}Pu	0.1077	0.0851
^{239}Pu	2.9618	2.8638
^{240}Pu	1.1531	1.1868
^{241}Pu	0.6246	0.5663
^{242}Pu	0.2562	0.2482
Total	5.1034	4.9502
^{143}Nd	0.3965	0.4461
^{144}Nd	0.6070	0.7428
^{145}Nd	0.3217	0.3835
^{146}Nd	0.3292	0.3923
^{148}Nd	0.1807	0.2099
^{150}Nd	0.0905	0.0991
Total	1.9256	2.2737

* calculated from ORIGEN2 by assuming that the specific gravity of HNO_3 solution before and after fuel dissolution not be changed

The presence of a mass 147 peak indicates the presence of either ^{147}Pm or ^{147}Sm. The ^{147}Sm/^{149}Sm isotopic ratio in naturally occurring Sm is 1.1, and the fission yield ratio for the mass 147 and mass 149 chains is 2.1. A measured mass 147/149 ratio greater than 2.1 indicate the presence of ^{147}Pm in the Nd fraction, and contribution to the mass 148 peak is ignored since ^{147}Pm is the only long-lived Pm isotope produced in the fission process. A mass 147/149 ratio approaching a value of 2.1 indicates the presence of Sm and that ^{148}Sm may impart a bias to the ^{148}Nd measurement. In case that the 147/149 ratio is large, any Sm isotope would not be present in the Nd fraction and therefore the ^{148}Sm contribution to the mass 148 peak can be ignored. From the mass spectrometric result of a Nd fraction separated from irradiated

fuel, the 147/149 ratio was calculated to be 397.5.

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. These values can be compared with those calculated from ORIGEN2 and sample preparation history(Table 3).

3.2. Isotope Correlations

The isotope correlation on the basis of the analysis of irradiated fuel samples is to express the dependence between individual isotopes of U, Pu and Nd. The mass spectrum of U, the mass 234, 235, 236, 238, can be observed, although the ^{234}U content is small. The Pu isotopes which characterize the burnt U fuel are ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu. However, the determination of ^{234}U and ^{238}Pu contents was suffered from a high error, so they were not considered for constructing the dependence. In our previous work[23], it was expressed that the dependences of uranium and plutonium isotopes on total burnup. The uranium isotopes were characterized by a decrease of ^{235}U and by an increase of ^{236}U and ^{238}U with increasing burnup. The percentage of ^{236}U and ^{238}U increases nearly linearly with the ^{235}U depletion. The low increase of ^{238}U percent content was characterized with increasing total burnup, which is obtain-

Table 4. Summary of Isotope Correlations for Irradiated PWR Fuel

No.	Isotope Correlation	Linear Dependence
1	at.% ^{235}U vs. at.% ^{236}U	at.% ^{236}U = -0.158 at.% ^{235}U + 0.563
2	$^{235}U/^{238}U$ vs. $^{236}U/^{238}U$	$^{236}U/^{238}U$ = -0.153 $^{235}U/^{238}U$ + 5.66 x 10^{-3}
3	D_5 vs. $^{235}U/^{238}U$	$^{235}U/^{238}U$ = -3.33 x 10^{-2} D_5 + 3.32 x 10^{-2}
4	D_5 vs. $^{236}U/^{238}U$	$^{236}U/^{238}U$ = 5.09 x 10^{-3} D_5 + 5.85 x 10^{-4}
5	D_5 vs. Pu/U*	Pu/U = 1.36 x 10^{-2} D_5 - 3.13 x 10^{-4}
6	at.% ^{235}U vs. Pu/U*	Pu/U = -4.21 x 10^{-3} at.% ^{235}U + 1.33 x 10^{-2}
7	at.% ^{236}U vs. Pu/U*	Pu/U = 2.69 x 10^{-2} at.% ^{236}U - 1.77 x 10^{-3}
8	$^{235}U/^{238}U$ vs. $^{240}Pu/^{239}Pu$	$^{240}Pu/^{239}Pu$ = -20.620 $^{235}U/^{238}U$ + 0.617
9	$^{235}U/^{238}U$ vs. F_t	F_t = -187.072 $^{235}U/^{238}U$ + 4.925
10	$^{236}U/^{238}U$ vs. F_t	F_t = 1207.099 $^{236}U/^{238}U$ - 1.930
11	$^{240}Pu/^{239}Pu$ vs. F_t	F_t = 8.955 $^{240}Pu/^{239}Pu$ - 0.626
12	$^{235}U/^{238}U$ vs. F_5	F_5 = -77.058 $^{235}U/^{238}U$ + 2.617
13	$^{236}U/^{238}U$ vs. F_5	F_5 = 493.841 $^{236}U/^{238}U$ - 0.193
14	$^{146}Nd/^{145}Nd$ vs. F_t	F_t = 11.375 $^{146}Nd/^{145}Nd$ - 8.226
15	$^{144}Nd/^{143}Nd$ vs. F_t	F_t = 3.049 $^{144}Nd/^{143}Nd$ - 1.995
16	$^{146}Nd/^{148}Nd$ vs. F_t	F_t = 18.735 $^{146}Nd/^{148}Nd$ - 31.987
17	$^{150}Nd/^{148}Nd$ vs. F_t	F_t = 34.201 $^{150}Nd/^{148}Nd$ - 13.137
18	$^{148}Nd/^{145}Nd$ vs. F_t	F_t = 28.181 $^{148}Nd/^{145}Nd$ - 11.875
19	$^{143}Nd/^{145+146}Nd$ vs. F_t	F_t = -10.873 $^{143}Nd/^{145+146}Nd$ + 9.560
20	$^{148}Nd/^{145+146}Nd$ vs. F_t	F_t = 138.081 $^{148}Nd/^{145+146}Nd$ - 33.649

F_t : Total burnup, F_5 : ^{235}U burnup, D_5 : ^{235}U Depletion, * : Mass ratio

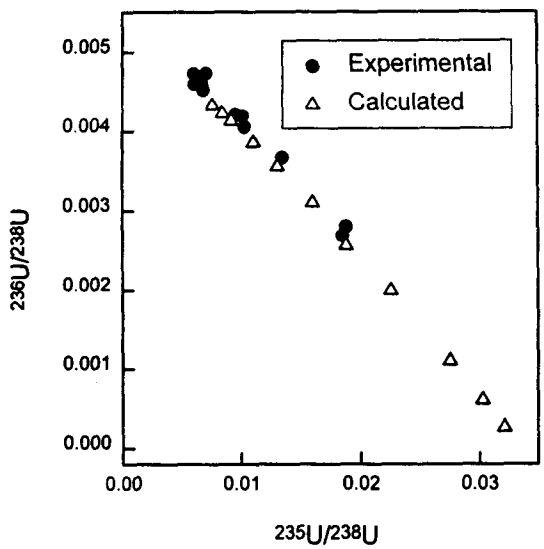

Fig. 2. Correlation Between $^{236}U/^{238}U$ and $^{235}U/^{238}U$ Atom Ratio

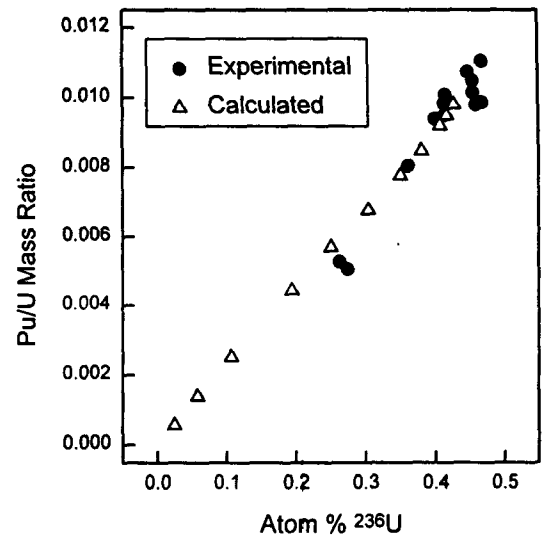

Fig. 3. Correlation Between Pu/U Mass Ratio and Atom % ^{236}U

ed by the Nd-148 method. The dependences of Pu composition on burnup were characterized by a decrease of ^{239}Pu percent content and an increase of ^{240}Pu, ^{241}Pu and ^{242}Pu percent contents.

In this paper, it is evaluated that the dependences of various isotope ratios for uranium, plutonium and neodynium against some parameters, e. g. total burnup, and correlations between isotope themselves, which are given Table 4. The dependences derived from experimental data are expressed by linear equations in Table 4.

3.2.1. Correlation between the Depletion, Pu/U Ratio and Heavy Atom Isotopes

The dependence of the concentration of ^{236}U on the depletion of ^{235}U expressed by equation 2 is shown in Fig. 2. This dependence shows that the $^{236}U/^{238}U$ atom ratio can be determined from the $^{235}U/^{238}U$ atom ratio. The dependence of ^{236}U and ^{235}U on ^{238}U can also be expressed in the form of atom % ^{236}U and atom % ^{235}U(equation 1). Occasionally it is more useful to correlate the depen-

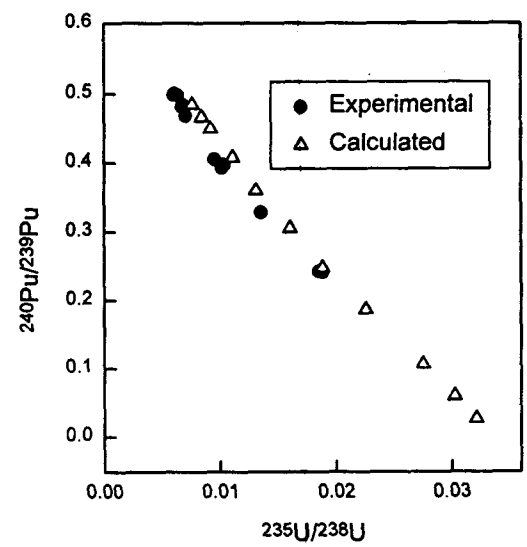

Fig. 4. Correlation Between $^{240}Pu/^{239}Pu$ and $^{235}U/^{238}U$ Atom Ratio

dence of the percent content of ^{236}U or ^{235}U, and $^{236}U/^{238}U$ or $^{235}U/^{238}U$ on the depletion of $^{235}U(D_5$, the difference between the initial and final ^{235}U content expressed in $W_5^o/W_5^o - W_5$) by equations 3 and 4. The equation 4 is used for the calculation of the ratio of the effective cross section for the activation

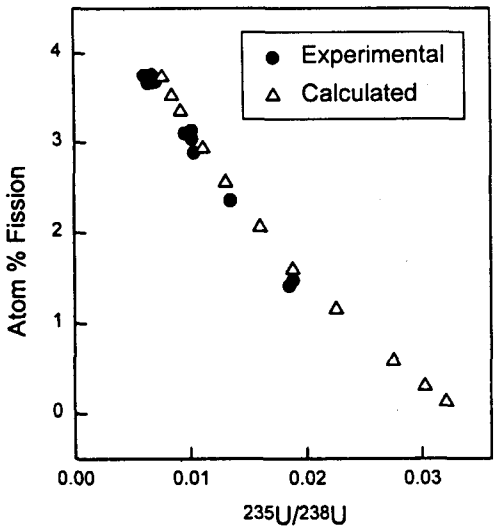

Fig. 5. Correlation Between Total Burnup(F_t) and $^{235}U/^{238}U$ Atom Ratio

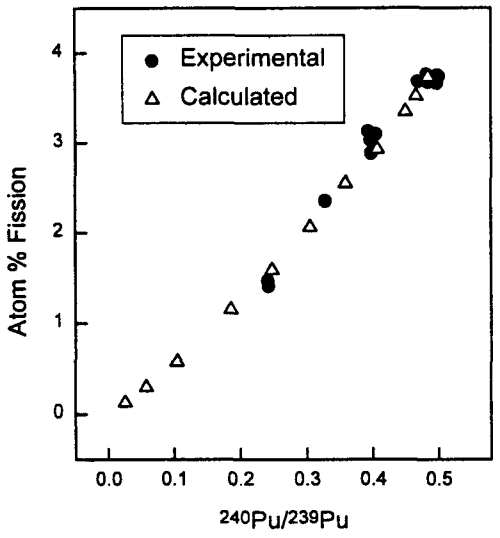

Fig. 6. Correlation Between Total Burnup(F_t) and $^{240}Pu/^{239}Pu$ Atom Ratio

and fission of ^{235}U, α_5[1].

The other correlations were expressed by the dependence between the content of the U isotopes and Pu content in the irradiated fuel. The correlations of Pu content on ^{235}U or ^{236}U percent content, and on ^{235}U depletion(D_5) derived from experimental data are expressed by equations 5, 6 and 7. The dependence of Pu/U mass ratio on ^{236}U percent content is shown in Fig. 3. From these dependences the Pu content can be calculated on the basis of known ^{235}U and ^{236}U percent contents. The correlation of the isotope composition of U and Pu determined experimentally, e.g. the $^{235}U/^{238}U$ ratio versus the $^{240}Pu/^{239}Pu$ expressed by equation 8 showed a good linearity(Fig. 4).

3.2.2. Correlation Between the Burnup and Heavy Atom Isotopes

The other correlations can be expressed against the total burnup(F_t) obtained by the Nd-148 method [23,24]. The correlation between Pu/U mass ratio and total burnup, and the dependences of U and Pu isotopes, e.g., ^{235}U, ^{236}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu on total burnup were shown in our previous work[23]. The dependences of the total burnup against $^{235}U/^{238}U$, $^{236}U/^{238}U$ and $^{240}Pu/^{239}Pu$ atom ratio determined experimentally are expressed by equations 9, 10 and 11, respectively. The dependences of total burnup against $^{235}U/^{238}U$ and $^{240}Pu/^{239}Pu$ ratio from experimental data are also shown in Figs. 5 and 6.

The ^{235}U burnup(F_5), which is fractional burnup from fission of ^{235}U, was measured by U and Pu mass spectrometric method[23,25]. The $^{235}U/^{238}U$ and $^{236}U/^{238}U$ atom ratio determined experimentally are expressed with good linearity against the ^{235}U burnup by equations 12 and 13, respectively.

3.2.3. Correlation Between the Burnup and Neodymium Isotopes

Since ^{148}Nd is commonly used as a monitor for the measurement of the number of fission and burnup, the Nd isotopes can be used easily for correlation at the same time with burnup measurement. Some correlation studies involving Nd isotopes have been reported[2,6,10,11]. The correlation of ^{143}Nd/

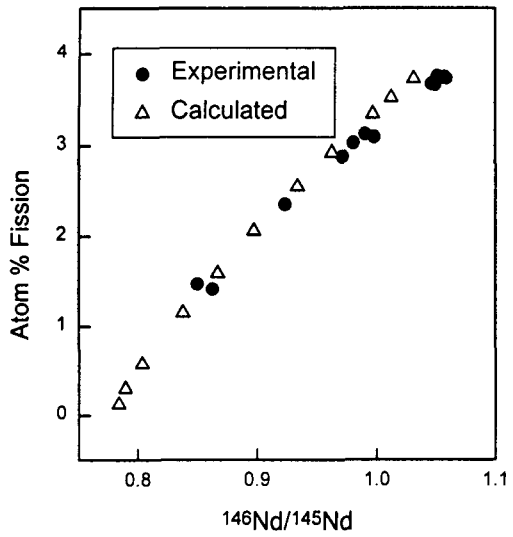

Fig. 7. Correlation Between Total Burnup(F_t) and ^{146}Nd / ^{145}Nd Atom Ratio

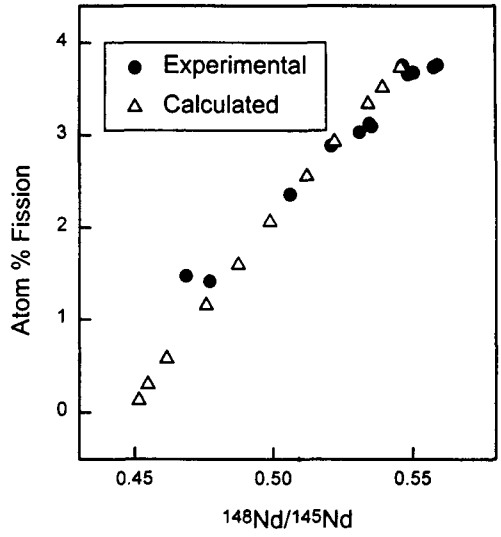

Fig. 8. Correlation Between Total Total Burnup(F_t) and $^{148}Nd/^{145}Nd$ Atom Ratio

$^{145+146}Nd$ with total burnup has wider applicability than any of the other correlations previously discussed. The use of the sum of ^{145}Nd and ^{146}Nd is preferred to ^{144}Nd because information relative to the decay of ^{144}Ce is not required[10]. Although the ratio of $^{145}Nd/^{146}Nd$ varies with burnup, the sum of $^{145+146}Nd$ isotopic fraction to total Nd is constant for all levels of burnup in irradiated fuel. Of particular importance is the absence of a significant neutron spectrum effect.

^{148}Nd has been used as a standard fission monitor, however, it has now been conceived that a large neutron capture cross section(~440 b) associated with $^{147}Nd(t_{1/2}=11$ d) can give rise to high biased values, especially when the sample has been exposed to a high neutron flux. The sum of ^{145}Nd and ^{146}Nd is therefore prefer to ^{148}Nd for correlation.

We expressed the correlations based on total burnup obtained by Nd-148 method and some Nd isotope ratios measured experimentally and then corrected for the contribution of contamination with natural Nd, e.g., $^{146}Nd/^{145}Nd$, $^{144}Nd/^{143}Nd$, $^{146}Nd/^{148}Nd$, $^{150}Nd/^{148}Nd$, $^{148}Nd/^{145}Nd$, $^{143}Nd/^{145+146}Nd$ and $^{148}Nd/^{145+146}Nd$. The isotope correlations of these Nd isotope ratios

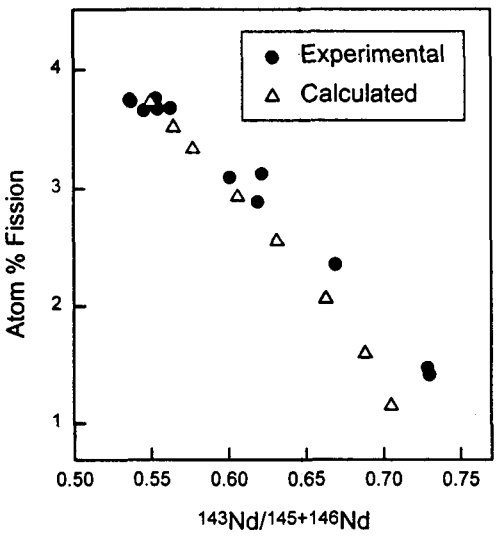

Fig. 9. Correlation Between Total Burnup(F_t) and ^{143}Nd / $^{145+146}Nd$ Atom Ratio

with total burnup showed a good linearity by equations 14 to 20, respectively. The correlations of $^{146}Nd/^{145}Nd$, $^{148}Nd/^{145}Nd$ and $^{143}Nd/^{145+146}Nd$ with total burnup are also shown in Figs. 7, 8 and 9. Therefore it is possible to determine the burnup of irradiated PWR fuel within a few percent of difference based

on only the Nd isotopic composition. However, the relation between the isotope ratio of $^{144}Nd/^{143}Nd$ and total burnup measured experimentally was different from that of calculation from ORIGEN2 code. These correlations between $^{144}Nd/^{143}Nd$ ratio and total burnup from the experimental and calculated results could not be illustrated by the same linear dependence. The $^{144}Nd/^{143}Nd$ ratios plotted against total burnup from the experimental results were about 1.5 to 2 times compared with the calculated values within the burnup range invegested. It seems that this disagreement of the experimental results with the calculated one is attributed to the accuracy of the nuclear data used in the calculation including the decay chain of ^{144}Ce-^{144}Pr-^{144}Nd and the neutron spectrum effect[2,10].

4. Conclusions

Several correlations of heavy elements and Nd isotope ratios with burnup values and/or some nuclear fuel parameters involved in burnup for irradiated PWR fuels Kori-1 reactor were evaluated. For practical use it will be necessary to establish the reliable correlations for each type of reactor and fuel characteristics(initial enrichment etc.) on the basis of the comparison of experimental and calculated results. Further correlation studies based on the other stable fission products and radioactive fission products will be performed in the future.

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