Burnup Determination of Dry Process Refabrication Fuel by Using Neodymium Isotope Monitors

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1. Introduction

One of the important parameters required for studies of irradiated nuclear fuel is burnup, which is the number of fission per 100 heavy nuclide atoms initially present in the fuel. Destructive method, which is based on the determination of specific nuclides by chemical analysis after appropriate separation procedures, is widely used as a reference method to measure the burnup of irradiated fuel[1]. 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. An 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 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[2,3]. 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 aim of this work is to determine the total burnup by using various Nd monitors on the same sample from SIMFUEL and dry process refabrication fuels irradiated in Hanaro reactor and to compare the results for the validity of the methods. In addition, the dependences of U, Pu and Nd isotope composition on burnup values, and correlations between U and Pu and their isotopes for these fuel samples were characterized experimentally.

2. Methods and Results

2.1 Irradiated Fuel Dissolution and Sample Preparation

A irradiated fuel sample weighed exactly was placed in a dissolution flask of dissolution apparatus. The fuel sample was refluxed for more than 10 hours in HNO_3(1+1) without catalyst. This fuel solution was weighed and an aliquot was diluted with the HNO_3(1+1) with the aid of ORIGEN calculation for the estimation of nuclides content in the irradiated 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.

2.2 Separation Procedure

Chemical separation was carried out for both the unspiked and the spiked(^{233}U, ^{242}Pu and ^{150}Nd) 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 reference [3,4].

2.3 Determination of Isotopic Composition

The U, Pu and Nd fractions collected from the spiked and unspiked fuel samples were prepared for mass spectrometric determination. In this work, all measured average ratios of Nd were corrected for mass discrimination to achieve high accuracy on burnup measurement. 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 1. Total Burnup Determined by Nd-148 Method for the Irradiated SIMFUEL and Dry Process Refabrication Fuel Samples

Fuel	MWD/MTHM			Irradiation
	Nd-148 (a)	Calc. (b)	Diff. (b/a)	
D4C1	41,798±1,306	42,200	1.010	Gori-1+Hanaro
D4C2	3,308±103	3,300	0.998	Hanaro
D4C3	31,154±974	30,600	0.982	Gori-1+Hanaro
D3C	39,869±1,246	38,900	0.976	Gori-1+Hanaro

D4C2 : SIMFUEL
b : Calculated from ORIGEN-2 code

2.4 Determination of Effective Fission Yield and Burnup

In this work, burnup value(in atom % fission) of the irradiated SIMFUEL and dry process refabrication fuel by various Nd isotope monitors was calculated by a procedure in reference[1]. Neodymium-148, 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 methods in reference[4,5]. Table 1 gives the total burnup(atom % fission) determined experimentally by Nd-148 method and calculated from ORIGEN-2 code for each fuel samples. The data in Table 1 were in agreement within 2.4%. Table 2 gives the total burnup measured by various monitors, ^{145}Nd + ^{146}Nd and ^{148}Nd isotopes. The data by two methods were in agreement within 1.3% for the same fuel sample.

Table 2. Comparison of Total Burnup Determined by Different Nd isotope Monitors for the Irradiated SIMFUEL and Dry Process Refabrication Fuel Samples

Fuel	MWD/MTHM			Irradiation
	Nd-148 (a)	Nd-(145+146) (b)	Diff. (b/a)	
D4C1	41,798	41,656	0.997	Gori-1+Hanaro
D4C2	3,308	3,264	0.987	Hanaro
D4C3	31,154	30,808	0.989	Gori-1+Hanaro
D3C	39,869	39,695	0.996	Gori-1+Hanaro

D4C2 : SIMFUEL

2.5 Correlation between Burnup and Isotope Compositions

In this work, It was evaluated that the dependences of various isotope ratios for U, Pu and Nd against burnup, and correlations between isotope themselves, e.g. ^{242}Pu/^{240}Pu and total burnup, ^{235}U/^{238}U and total burnup, ^{236}U/^{238}U and total burnup, ^{235}U/^{238}U and ^{236}U/^{238}U, atom % U isotope and total burnup(Figure 1), atom % Pu isotope and total burnup(Figure 2), ^{148}Nd/^{145}Nd and total burnup, ^{144}Nd/^{143}Nd and total burnup, ^{143}Nd/$^{145+146}$Nd and total burnup, atom % ^{235}U and ^{143}Nd/$^{145+146}$Nd, atom % ^{239}Pu and ^{143}Nd/$^{145+146}$Nd. The isotope ratios determined experimentally were expressed with good linearity against the total burnup and isotope compositions from experimental data.

3. Conclusion

The use of all the Nd isotopes in the determination of burnup for nuclear fuel has the advantage of confirming the value obtained for ^{148}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. The Nd isotope pattern provides information on the real irradiation characteristics which is necessary for evaluating the fuel performance in the reactor.

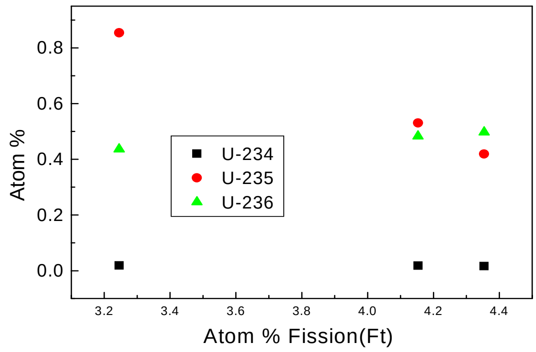

Figure 1. The Dependence of U Isotopes on Total Burnup(Ft).

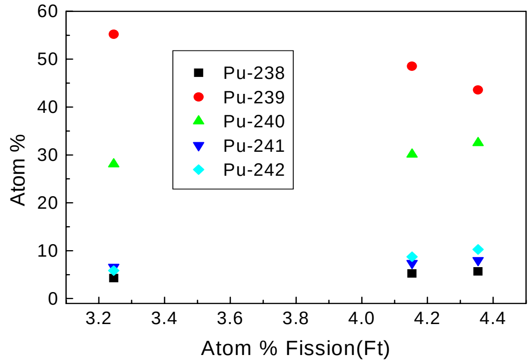

Figure 2. The Dependence of Pu Isotopes on Total Burnup(Ft).

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