Burnup Determination of Irradiated Sintered Solid and Annular Pellets by Neodymium Monitor Methods Based on Isotope Dilution Mass Spectrometric Measurements

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1. Introduction

One of the important parameters required for the study of a nuclear fuel is its burnup, which is the number of fission per 100 heavy nuclide atoms (mass ≥232) initially present in the fuel. Destructive methods, which is based on the determination of specific nuclides by a chemical analysis after appropriate separation of the heavy elements and a monitoring fission product, are 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. However, a serious drawback of ^{148}Nd burnup monitor is its reported susceptibility to a ^{147}Nd neutron capture effect, that is, a large thermal neutron capture cross- section for ^{147}Nd which would cause increasing amounts of ^{148}Nd with increasing neutron flux. The isotope ^{142}Nd, meanwhile, is used to monitor and correct possible contamination of the fission product with natural Nd during chemical separation. Another approach is to use a different monitor such as ^{145}Nd +^{146}Nd because the sum of ^{145}Nd and ^{146}Nd appears to be invariant with neutron flux and fluence[2]. 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 neodymium have to be added.

The aim of the present work is to determine the total burnup by using various neodymium isotope monitors for the same samples from the sintered solid and dual-cooled annular pellets irradiated in the Hanaro reactor at KAERI, and is to compare the results, so as to determine the respective validity of the methods.

2. Experiments

2.1 Chemicals

Certified ^{233}U (99.470 atom%) and ^{150}Nd (96.13 atom%) spikes were obtained from Oak Ridge National Laboratory (ORNL). Certified ^{242}Pu spike solution (99.9033 atom%, IRMM-044) was obtained from the Institute for Reference Materials and Measurements.

2.2 Sample Preparation and Isotopic Measurement

The irradiated pellet sample having been precisely weighed was placed into a 100 mL dissolution flask of the dissolution apparatus. 30~40 mL of 8 M HNO_3 for the samples of S1, S5, S6 and S7, and 30~40 mL of 8 M HNO_3 and 3 mL of c-HCl for the samples of S2, S3 and S4, which contain some additives such as Al, Mn and Cr, was then added, while applying a water cooling. All samples was refluxed for more than 12 hours. Chemical separation was carried out for both the unspiked and the spiked sample solutions in the same experimental conditions in a glove box. The isotopic compositions of U, Pu and Nd in the unspiked and spiked samples were determined using a thermal ionization mass spectrometer (TIMS, Finnigan TRITON).

3. Results & Discussion

3.1 Determination of the Isotopic Composition

Table 1 shows the isotopic compositions of neodymium in the irradiated pellet samples measured by the TIMS. In this work, all the measured average ratios of neodymium were corrected for a mass discrimination and the contribution of a natural contamination, so as to achieve a high accuracy for the burnup measurement.

3.2 Determination of the Total burnup and Burnup Parameters

Table 2 gives the total burnup in GWd/MtU determined by the neodymium isotope monitors, that is, ^{148}Nd, ^{143}Nd+^{144}Nd, ^{145}Nd+^{146}Nd, and the total of the Nd isotopes for the irradiated pellet samples. The number of fissions by the ^{148}Nd monitor was calculated with a correction for the excess ^{148}Nd produced from the capture on ^{147}Nd[1]. The data obtained by using other neodymium isotope monitors are in agreement, within a deviation of 5%, with those by the ^{148}Nd monitor. Table 3 shows the F_5 (^{235}U fractional burnup) and D_5 (^{235}U depletion) values, ^{148}Nd/U and Nd/U mass ratios. The F_5 is calculated by assuming no loss of ^{238}U or ^{236}U during an irradiation. The D_5 (the difference between the initial and final ^{235}U content) is expressed by $W_5^O/(W_5^O-W_5)$ [2].

4. Conclusion

The contents of U, Pu, Nd and their isotopes in irradiated sintered solid and annular pellet samples and the total burnup by using various neodymium isotope monitors can be determined simultaneously by the isotope dilution mass spectrometric techniques. The neodymium isotope patterns provide information on the real irradiation characteristics which are necessary for evaluating a fuel's performance in a reactor. A comparison between independently determined burnup values provides a check on the validity of the results

Table 1. Isotopic Compositions of the Neodymium Separated from the Irradiated Pellet Samples

	Atom%*					
	^{143}Nd	^{144}Nd	^{145}Nd	^{146}Nd	^{148}Nd	^{150}Nd
S1	10.35	42.31	16.21	17.74	9.31	4.08
S2	10.98	41.72	16.35	17.60	9.28	4.07
S3	11.09	41.72	16.36	17.57	9.23	4.03
S4	10.69	42.04	16.27	17.68	9.27	4.04
S5	10.20	42.58	16.10	17.75	9.29	4.08
S6	9.84	42.73	15.99	17.92	9.39	4.14
S7	9.87	42.70	16.00	17.86	9.40	4.16

* Corrected for contribution due to natural contamination and mass discrimination.

Table 2. Total Burnup Determined by the Neodymium Isotope Monitor Methods for the Irradiated Pellet Samples

	GWd/MtU			
	^{148}Nd	^{143}Nd+^{144}Nd	^{145}Nd+^{146}Nd	Nd-total
S1	33.28 ±1.04	34.21 ±1.07	34.32 ±1.07	33.53 ±1.05
S2	33.22 ±1.04	33.47 ±1.05	33.90 ±1.06	33.18 ±1.04
S3	33.20 ±1.04	33.31 ±1.04	33.80 ±1.06	33.05 ±1.03
S4	34.00 ±1.06	34.34 ±1.07	34.78 ±1.09	33.95 ±1.06
S5	35.45 ±1.11	35.81 ±1.12	36.23 ±1.13	35.40 ±1.11
S6	36.43 ±1.14	36.84 ±1.15	37.31 ±1.17	36.39 ±1.14
S7	38.83 ±1.21	36.87 ±1.15	37.28 ±1.17	37.18 ±1.16

Table 3. Determination of Some Burnup Parameters for the Irradiated Pellet Samples

	Atom%*				
	^{148}Nd/U	Nd/U	D_5	F_5	F_5/F_T
S1	4.02E-4	4.23E-3	1.033	2.244	0.647
S2	3.94E-4	4.16E-3	1.039	2.223	0.642
S3	3.89E-4	4.13E-3	1.039	2.226	0.644
S4	4.02E-4	4.25E-3	1.035	2.234	0.631
S5	4.19E-4	4.42E-3	1.030	2.244	0.608
S6	4.35E-4	4.54E-3	1.025	2.258	0.595
S7	4.36E-4	4.54E-3	1.025	2.257	0.558

D_5: ^{235}U depletion in weight, F_5 : ^{235}U fractional burnup in atom% fission, F_T : total burnup in atom% fission

REFERENCES

[1] ASTM, "Standard Test Method for Atom Percent Fssion in Uranium and Plutonium Fuel (Neodymium-148 Method)", Annual Book of ASTM Standards 12.02, E321-96 (reapproved 2012), (2012).

[2] J. S. Kim, Y. S. Jeon, S. D. Park, S. H. Han, and J. G. Kim, "Burnup Determination of High Burnup and Dry Processed Fuels Based on Isotope Dilution Mass Spectrometric Measurements", J. Nucl. Sci. & Technol., 44(7) 1015-1023 (2007).