Determination of Cerium Isotopes and Total Burnup in Irradiated Pellet Based on Isotope Dilution Mass Spectrometric Measurement

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1. Introduction

The burnup of important fissile isotopes and the composition of an irradiated fuel depend on the nature of the fuel and on the conditions of an irradiation. A detailed knowledge of these quantities is useful for a reactor work as well as for the effective utilization of a nuclear fuel. Burnup determination by destructive method, which is based on the determination of specific nuclides, e.g. U, Pu and ^{148}Nd by a chemical analysis after an appropriate separation of the heavy elements and a monitoring of the fission product, is widely used as a reference method to measure the burnup of an irradiated fuel [1]. The ^{140}Ce and ^{142}Ce were recommended as useful nuclear fuel burnup monitors of total fission because they satisfy most of the necessary requirements for a good burnup monitor of PWR fuel, and can be determined easily using isotope dilution mass spectrometric techniques (IDMS) [2].

The aim of the present work is to determine the isotopic compositions and its contents of Ce for the samples from sintered solid and annular pellets of PWR type irradiated in the Hanaro reactor at KAERI, and to determine the total burnup using the measured results, so as to determine the respective validity of the methods

2. Experiments

2.1 Chemicals

The Certified ^{140}Ce (99.84 atom%) spike was obtained from Oak Ridge National Laboratory (ORNL). This spike solution was prepared by dissolving its oxide in hot c-HNO_3 added H_2O_2. The concentration of the spike solution was determined by calibrating that with a Ce standard solution and inductively coupled plasma–atomic emission spectrometer (ICP-AES).

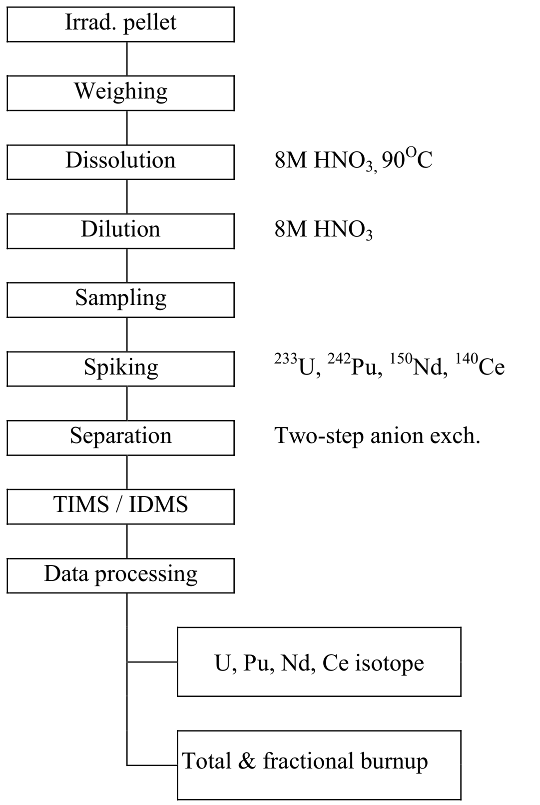

Fig. 1. Analytical processes for the determination of U, Pu, Nd and Ce isotopes, and burnup in an irradiated pellet.

2.2 Chemical Separation and Isotopic Determination

The basic processes in the analytical laboratory for the determination of U, Pu, Nd, Ce, and burnup are shown in Fig. 1. Chemical separation was carried out for both the unspiked and the spiked sample solutions in the same experimental conditions in a glove box without any heavy shieldings. Nd is eluted from other fission products on the second anion exchange resin column (AG 1X4, 200-400 mesh) with 0.04M HNO_3-99.8% MeOH (1:9) eluent, followed by Ce elution with 0.007M HNO_3-99.8% MeOH (1.5:8.5) eluent. The isotopic compositions of U, Pu, Nd and Ce in the unspiked and spiked pellet samples were determined using a thermal ionization mass spectrometer (TIMS, Finnigan TRITON).

2.3 Determination of the Ce and total burnup

The concentrations of Ce in the sample solutions were determined by the IDMS according to Eq. (1):

$$C_n = C_a \cdot G_a/G_n \cdot M_n/M_a \cdot (R_a-R_m)/(R_m-R_n) \cdot (\sum nRi)/(\sum aRi) \quad \text{------ (1)}$$

Where,
C_n : concentration of Ce in sample soln. (μg-Ce/ mL)
C_a : concentration of Ce in spike soln. (μg-Ce/mL)
G_n : volume of sample soln. taken (mL)
G_a : volume of spike soln. taken (mL)
M_n : mean atomic weight of Ce in sample
M_a : mean atomic weight of Ce in spike
R_a : ratio of two basic isotopes in spike ($^{142}Ce/^{140}Ce$)
R_m: ratio of two basic isotopes in mixture($^{142}Ce/^{140}Ce$)
R_n : ratio of two basic isotopes in sample ($^{142}Ce/^{140}Ce$)
∑nRi : sum of ratios of total isotopes for basis isotope in sample
∑aRi : sum of ratios of total isotopes for basis isotope in spike

The total burnup in atom% fission for the pellet sample was determined by using Nd and Ce isotope monitors according to Eq. (2) [3]:

$$\text{Atom\% fission} = (N/Y) \times 100 / [(N/Y) + (N/\text{U}) + (N/\text{Pu})] \quad \text{------ (2)}$$

Where,
N : number of atoms of the monitor Nd or Ce isotope in the sample solution,
Y : effective fission yield of the monitor Nd or Ce isotope from the fissile elements,
N(U), *N*(Pu) : number of U and Pu atoms in the sample solution , respectively.

3. Results & Discussion

3.1 Isotopic Compositions of Ce in Irradiated Fuel

The Ce isotopes in irradiated pellets include two major ^{140}Ce and ^{142}Ce, and trace ^{144}Ce (ɣ-emitter). Table 1 shows the isotopic compositions of Ce in the irradiated pellet samples (B3 and BO3), and their spiked samples measured by the TIMS .

3.2 Determination of the effective fission yield

The effective fission yields of the Ce isotope monitors were calculated from the fission yields for each of the fissioning isotopes weighted according to their contribution to the fission (Table 2). The concentration of fissionable isotopes used those determined by the IDMS.

Table 1. Isotopic compositions of the Ce separated from the irradiated pellet samples

	Atom%		
Sample	^{140}Ce	^{142}Ce	^{144}Ce
B3 (S)	53.225	46.308	0.467
BO3 (S)	53.123	46.380	0.497
B3 (S+SP)	73.900	25.841	0.259
BO3 (S+SP)	76.170	23.605	0.225
S : sample, S+SP : spiked sample			

Table 2. Estimated effective fission yields of Ce isotopes for the irradiated pellet samples

Isotope	Effective fission yield (%)	
	B3	BO3
^{140}Ce	5.9219	5.8898
^{142}Ce	5.4830	5.4400

4. Conclusion

The contents of U, Pu, Nd, Ce and their isotopes in irradiated pellet samples and the total burnup by using Nd and Ce isotope monitors can be determined simultaneously by the isotope dilution mass spectrometric techniques. The Nd and Ce 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

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