Nucl Eng Technol 47 (2015) 924–933

Available online at www.sciencedirect.com

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journal homepage: http://www.journals.elsevier.com/nuclear-engineering-and-technology/

Technical Note

ANALYSIS OF HIGH BURNUP PRESSURIZED WATER REACTOR FUEL USING URANIUM, PLUTONIUM, NEODYMIUM, AND CESIUM ISOTOPE CORRELATIONS WITH BURNUP

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ARTICLE INFO

Article history:
Received 13 March 2015
Received in revised form
28 July 2015
Accepted 9 August 2015
Available online 22 October 2015

Keywords:
Burnup determination
High burnup pressurized water reactor fuel
Isotope dilution mass spectrometric method
Isotopic correlation

ABSTRACT

The correlation of the isotopic composition of uranium, plutonium, neodymium, and cesium with the burnup for high burnup pressurized water reactor fuels irradiated in nuclear power reactors has been experimentally investigated. The total burnup was determined by Nd-148 and the fractional ^{235}U burnup was determined by U and Pu mass spectrometric methods. The isotopic compositions of U, Pu, Nd, and Cs after their separation from the irradiated fuel samples were measured using thermal ionization mass spectrometry. The contents of these elements in the irradiated fuel were determined through an isotope dilution mass spectrometric method using ^{233}U, ^{242}Pu, ^{150}Nd, and ^{133}Cs as spikes. The activity ratios of Cs isotopes in the fuel samples were determined using gamma-ray spectrometry. The content of each element and its isotopic compositions in the irradiated fuel were expressed by their correlation with the total and fractional burnup, burnup parameters, and the isotopic compositions of different elements. The results obtained from the experimental methods were compared with those calculated using the ORIGEN-S code.

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1. Introduction

To check the consistency of postirradiation analysis results, for example, mass spectrometric analyses, analytical techniques for chemical measurements, and information regarding reactor parameters and irradiation conditions, correlations between the parameters of irradiated nuclear fuels such as the concentrations of heavy elements and fission products, ratios of their isotopes, and total and fractional burnups were established [1–16]. Nuclear fuel in a reactor undergoes a variation in its isotopic composition, that is, the depletion of the fissile isotopes initially present, buildup of heavy elements, and buildup of fission products. The intrinsic coherence of isotopic abundances in irradiated fuels is

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http://dx.doi.org/10.1016/j.net.2015.08.002
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described by correlations observed not only between different isotopic ratios but also between isotopic ratios and burnup parameters. These correlations can therefore be used to check the consistency of the analytical results, and can also be used to identify reactor fuels and to estimate the burnup and Pu production. Some of these correlations may also be useful as safeguards. The isotopic correlations can be divided into three groups, namely, correlation based on heavy isotopes and elements, stable fission products, and radioactive fission products. Among the elements used in these studies, U, Pu, Nd, Cs, Eu, Ru, Zr, Kr, and Xe should be mentioned.

The parameters generally used for correlation studies are as follows: total burnup (F_t), ^{235}U fractional burnup (F_5), ^{235}U depletion (D_5), capture to fission ratio of ^{235}U (α_5), ^{239}Pu buildup, Pu/U ratio, U/U_O (where U_O is the initial U content) ratio, the fission contributions of ^{235}U, ^{239}Pu, and ^{241}Pu, and some fission product and heavy-element isotopic ratios in the irradiated fuel.

In this work, the dependencies of U, Pu, Nd, and Cs isotopic compositions on the burnup and other parameters, and the correlations between these element isotopes for the pressurized water reactor (PWR) uranium dioxide fuels irradiated to a high burnup in nuclear power reactors were experimentally characterized. The isotopic compositions of U, Pu, Nd, and Cs were determined by thermal ionization mass spectrometry and gamma-ray spectrometry, and the concentrations of those elements were determined using the isotope dilution mass spectrometric (IDMS) method using ^{233}U, ^{242}Pu, ^{150}Nd, and ^{133}Cs as spikes.

2. Experimental analysis

2.1. Chemicals and instruments

Certified ^{233}U (99.470 atom%) and ^{150}Nd (96.13 atom%) spikes were obtained from Oak Ridge National Laboratory (Oak Ridge, Tennessee, USA). These spike solutions were prepared by dissolving their oxides in 8M HNO_3–0.01M HF. A certified ^{242}Pu spike solution (99.9033 atom%, IRMM-044) was obtained from the Institute for Reference Materials and Measurements (Retieseweg, Geel, Belgium) (Table 1). The ^{133}Cs spike solution was obtained using monoisotopic standard solutions from Spex Industries Inc. (Metuchen, New Jersey, USA). NBL CRM 129 U_3O_8 powder (New Brunswick Laboratory; Argonne, Illinois, USA) and a U standard solution (Spex Industries Inc.) were used as standard reference materials for the uranium. Neodymium standard solutions were obtained from Accu-Trace (New Haven, Connecticut, USA) and Spex Industries Inc. The concentrations of spike solutions are as follows: 1,320.0712 μg ^{233}U/mL, 1.0706 μg ^{242}Pu/mL, 1.8922 μg ^{150}Nd/mL, and 2.0000 μg ^{133}Cs/mL. The isotopic compositions of U, Pu, Nd, and Cs in the unspiked and spiked samples were determined using a thermal ionization mass spectrometer of the Finnigan MAT 262 type. The activity of gamma emitters for the samples diluted from the dissolved solutions was measured with a high-purity Ge coaxial detector (EG & G ORTEC) connected to a multichannel analyzer. The PWR nuclear fuels used in this work were irradiated to a high burnup of between 34 GWd/MtU and 62 GWd/MtU in three power reactors (arbitrarily, A, B, and C) in Korea for 1,200–1,400 days (EFPDs, effective full-power days), with various enrichments of 4.2–4.5 w/o, and then cooled for about 2 years for postirradiation analyses.

Table 1 – Isotopic compositions of the certified spikes.

Spike	Isotope	Atom%
	U-233	99.470
	U-234	0.166
^{233}U	U-235	0.064
	U-236	0.015
	U-238	0.282
	Pu-238	0.0009
	Pu-239	0.0826
	Pu-240	0.0108
^{242}Pu	Pu-241	0.0009
	Pu-242	99.9033
	Pu-244	0.0015
	Nd-142	0.77
	Nd-143	0.39
	Nd-144	0.88
^{150}Nd	Nd-145	0.34
	Nd-146	0.84
	Nd-148	0.66
	Nd-150	96.13
^{133}Cs	Cs-133	100.00

2.2. Irradiated fuel dissolution and sample preparation

The basic processes used in the PIE (post-irradiation examination) analytical laboratory for the burnup and isotopic determination are shown in Fig. 1. In a chemical hot cell, the

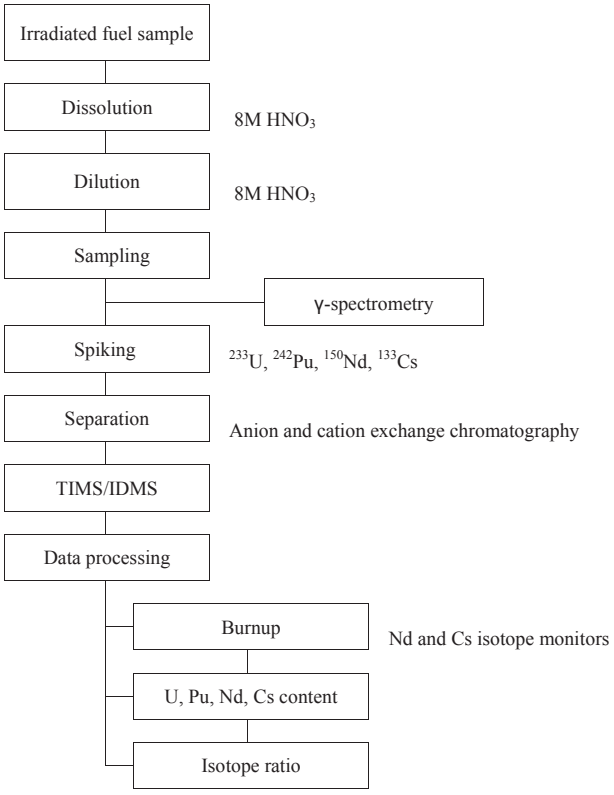

Fig. 1 – Basic processes in the PIE analytical laboratory. IDMS, isotope dilution mass spectrometry; PIE, post-irradiation examination:TIMS, thermal ionization mass spectrometer.

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