Determination of Uranium, Plutonium, Neodymium, Cesium, Cerium and Europium Isotopes in Irradiated Dual-cooled Annular Fuel by Isotope Dilution Mass Spectrometry and γ-Ray Spectrometry

Jung Suk Kim*, Byungman Kang, Yang-Soon Park, Young Bok Kim, Jai Il Park, and Yeong Keong Ha

Korea Atomic Energy Research Institute, 111, Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, Republic of Korea

*njskim1@kaeri.re.kr

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 (or ^{137}Cs) 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 [1-3]. Of the many stable nuclides formed in the fission, Nd satisfies most of the necessary requirements for a good burnup monitor[1], can be measured using isotope dilution mass spectrometric techniques (IDMS). On the other hand, radioactive nuclides such as ^{134}Cs, ^{137}Cs and ^{144}Ce can be measured using γ-ray spectrometric techniques (GRS).

The aim of the present work is to determine the isotopic compositions and their contents of U, Pu and Nd, and the activities of Cs, Ce and Eu isotopes (γ-emitters) for the samples from dual-cooled annular nuclear fuels of UO_2 type irradiated in the Hanaro reactor at KAERI, and to determine the burnup using the measured results, so as to determine the respective validity of the methods

2. Experiments

2.1 Chemicals

The Certified ^{233}U (99.470 atom%) and ^{150}Nd (96.13 atom%) spikes were obtained from Oak Ridge National Laboratory (ORNL). These spike solutions were prepared by dissolving their oxides in 8 M HNO_3-0.01 M HF. Certified ^{242}Pu spike solution (99.9033 atom%, IRMM-044) was obtained from the Institute for Reference Materials and Measurements. The concentrations of the spike solutions were prepared by calibrating them with a standard solution.

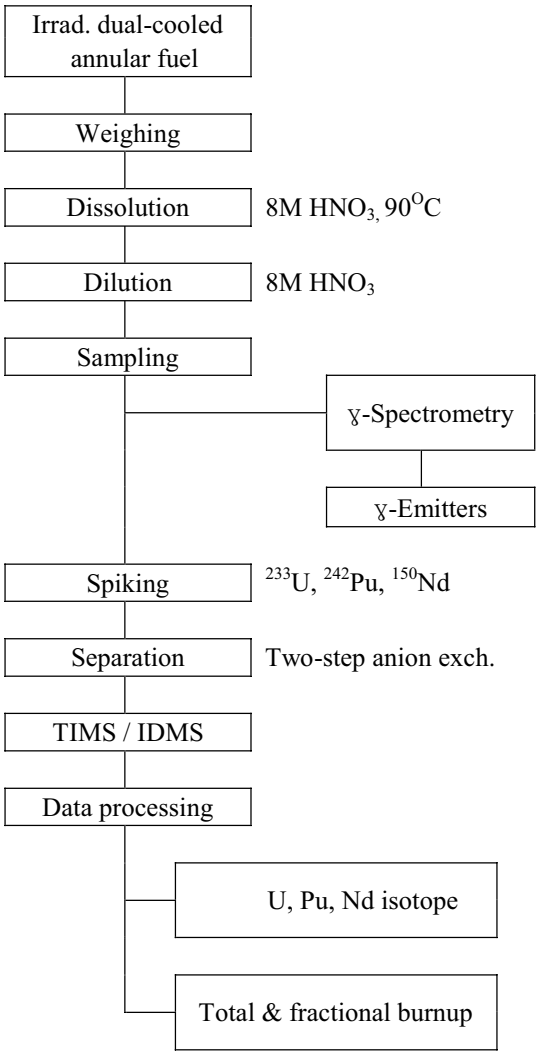

Fig. 1. Analytical Processes for the Determination of U, Pu, Nd Isotopes and γ-Emitters in an Irradiated Dual-cooled Annular Fuel Sample.

2.2 Sample Preparation and Isotopic Measurement

The basic processes in the analytical laboratory for the determination of U, Pu, Nd, γ-emitters, 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. The isotopic compositions of U, Pu and Nd in the unspiked and spiked fuel samples were determined using a thermal ionization mass spectrometer (Finnigan TRITON). The activity of gamma emitters for the diluted solutions from the fuel solutions was measured with a high purity Ge coaxial detector (EG & G ORTEC) connected to a multi-channel analyzer.

3. Results & Discussion

3.1 γ-Emitters in Irradiated Fuel

The activities of γ-emitters found in the dissolved solutions of irradiated dual-cooled annular fuels are summarized in Table 1. The activities of γ-emitters in the dissolved solutions from the GRS, that is, S-1, S-2 and S-3, include the fission products ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{154}Eu, ^{155}Eu, ^{106}Ru and ^{125}Sb and activation products such as ^{60}Co and ^{95}Zr.

Table 1. Distribution of Radionuclides in Irradiated Dual-cooled Annular Fuel Samples

Nucl.	Activity (Bq/g-sample)		
	S-1	S-2	S-3
^{134}Cs	1.58×10^{9} $\pm6.27\times10^{7}$	1.83×10^{9} $\pm7.23\times10^{7}$	1.76×10^{9} $\pm6.96\times10^{7}$
^{60}Co	2.17×10^{7} $\pm1.03\times10^{6}$	1.99×10^{7} $\pm1.00\times10^{6}$	2.71×10^{7} $\pm1.39\times10^{6}$
^{137}Cs	3.54×10^{9} $\pm1.44\times10^{8}$	3.48×10^{9} $\pm1.41\times10^{8}$	3.74×10^{9} $\pm1.53\times10^{8}$
^{144}Ce	2.91×10^{9} $\pm9.36\times10^{7}$	2.82×10^{9} $\pm9.23\times10^{7}$	3.00×10^{9} $\pm9.88\times10^{7}$
^{154}Eu	1.37×10^{8} $\pm6.58\times10^{6}$	1.44×10^{8} $\pm6.52\times10^{6}$	1.50×10^{8} $\pm7.10\times10^{6}$
^{155}Eu	5.97×10^{7} $\pm2.98\times10^{6}$	6.39×10^{7} $\pm3.11\times10^{6}$	7.35×10^{7} $\pm3.62\times10^{6}$
^{106}Ru	3.06×10^{9} $\pm2.06\times10^{8}$	3.10×10^{9} $\pm2.01\times10^{8}$	2.90×10^{9} $\pm1.95\times10^{8}$
^{125}Sb	1.21×10^{8} $\pm7.20\times10^{6}$	1.23×10^{8} $\pm8.53\times10^{6}$	1.46×10^{8} $\pm9.19\times10^{6}$
^{95}Zr	1.45×10^{7} $\pm2.57\times10^{6}$	1.11×10^{7} $\pm2.21\times10^{6}$	1.96×10^{7} $\pm4.59\times10^{6}$
^{140}Ba	$< 4.63\times10^{6}$	$< 7.33\times10^{6}$	$< 8.35\times10^{6}$
^{95}Nb	$< 4.42\times10^{6}$	$< 3.21\times10^{6}$	$< 7.38\times10^{6}$

3.2 Isotopic Compositions of Cs and Eu

Table 2 shows a comparison of the activity ratios for Cs and Eu in the the irradiated fuel samples, It may be inferred that there is a distinct difference between the three values with the progression of the fuel burnup.

Table 2. Comparison of Cs and Eu Activity Ratios in Irradiated Dual-cooled Annular Fuel Samples

Sample	Activity ratio	
	^{134}Cs/^{137}Cs	^{154}Eu/^{137}Cs
S-1	0.446	0.039
S-2	0.525	0.041
S-3	0.470	0.040

4. Conclusion

The contents of U, Pu, Nd, Cs and their isotopes in irradiated dual-cooled annular fuel samples and the total burnup by using Nd, Cs and Ce isotope monitors can be determined simultaneously by the isotope dilution mass spectrometric and γ-ray spectrometric techniques. The Nd and Cs 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

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, Y. K. Ha, and K.. Song, "Analysis of High Burnup Pressurized Water Reactor Fuel Using Uranium, Plutonium, Neodymium, and Cesium Isotope Correlations with Burnup", Nucl. Eng. & Technol., 47(7) 924-933 (2015).

[3] H. Graber, G. Hofmann and R. Berndt, "Correlations of Burnup with the ^{134}Cs/^{137}Cs and ^{154}Eu/^{137}Cs Concentration Ratios for Irradiated LWR Fuel", IAEA-SM-260/84, Nuclear Safeguards Technology, vol. I, 443-454 (1983).