OVERVIEW OF THE ANALYTICAL PROCESSES USED TO SIMPLIFY ATOM PERCENT FISSION ANALYSIS IN URANIUM AND PLUTONIUM FUELS BY ASTM E321-96

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Overview of the Analytical Processes Used to Simplify Atom Percent Fission Analysis in Uranium and Plutonium Fuels by ASTM E321-96

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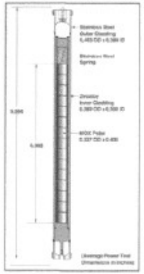
The Radioactive Materials Analytical Laboratory (RMAL) has performed measurements for atom percent fission in irradiated mixed oxide (MOX) fuel for the Department of Energy's Fissile Materials Disposition Program (FMDP) since 1999

To date four irradiation cycles have been examined and analyzed:

- 7.5 GWd/MT (Summer 1999)
- 22 GWd/MT (Spring 2000)
- 30 GWd/MT (Summer 2001)
- 40 GWd/MT (Winter 2003)

With each successive campaign, the processes involved in the percent atom fission determination have been improved and refined such that, for the final campaign coming up this summer (2004), the updated processes employed will be both effective and efficient, requiring a minimal amount of personnel dose and having much improved turnaround times compared with those of the initial campaign

FMDP Test Assembly



Summary of Analytical Procedure for Burnup Determinations

Irradiated fuel pellets are dissolved, and the resultant digests undergo separations for neodymium, plutonium, and uranium isotopes. The purified aliquots are quantified for their element of interest by isotope dilution mass spectrometry (IDMS)

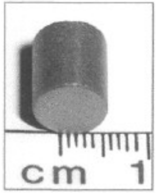
The measured IDMS concentrations for Nd, Pu, and U and their respective isotopic measurements are used to calculate each isotope's atom fraction (atom/g)

The percent burnup is calculated as the sum of the ^{235}U + ^{239}Pu atom fractions consumed (calculated as a direct ratio of the measured ^{148}Nd (atoms/g)/^{148}Nd effective yield from fission) divided by the initial total fissionable isotopes concentration ($^{235}U + ^{238}U + ^{239}Pu + ^{240}Pu + ^{241}Pu$). A simple conversion factor can be applied to convert units to GWd/MT

Sample Preparations

All sample digestions were performed in shielded hot cells on a hot plate with covered beakers at 150°C surface temperature

Sample sizes ranged from 2.5 to 5.0 g and consisted of whole irradiated pellets or pellet fragments



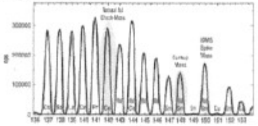
Unirradiated MOX Fuel Pellet

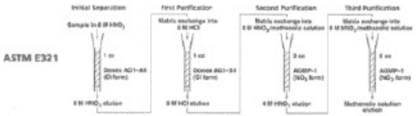
Irradiation Cycle	Acids Used	Time Needed	Digestion Comments
7.5 GWd/MT	HNO_3/HCl/HF	2 weeks	Saturated boric acid solution added to digestion to complex fluorides. Difficult matrix with large amounts of boron
22 GWd/MT	HNO_3/HCl	2 weeks	Trouble getting all solids completely in solution. Matrix adjustment required for separations
30 GWd/MT	HNO_3/HCl	2 weeks	Trouble getting all solids completely in solution. Matrix adjustment required for separations
40 GWd/MT	HNO_3	3 days	Very small amount of solids that remained in some samples dissolved when digestions were diluted to volume. Clean matrix

Even with increasing irradiation periods nitric acid alone is sufficient to completely dissolve the fuel pellets resulting in cleaner digestion solutions

Neodymium Separations

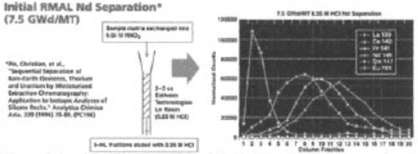
High-mass fission products scan of a digested MOX fuel pellet





ASTM separation scheme is time-consuming and requires a large amount of sample handling and a multitude of reagents

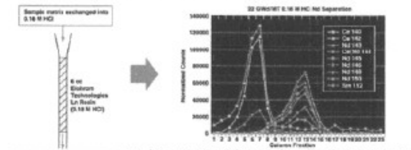
Initial RMAL Nd Separation* (7.5 GWd/MT)



*Pin, Christian, et al., "Sequential Separation of Rare-Earth Elements, Thorium and Uranium by Miniaturized Extraction Chromatography: Application to Isotopic Analyses of Silicate Rocks," Analytica Chimica Acta, 339 (1994) 79-89.

Near co-elution of the Nd isotopes with interfering Ce isotopes but baseline separation from the Sm isotopes

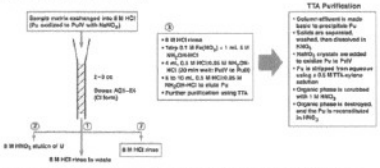
Refined RMAL Nd Separation (20, 30, and 40 GWd/MT)



Baseline separation of the Nd isotopes from the interfering Ce and Sm isotopes

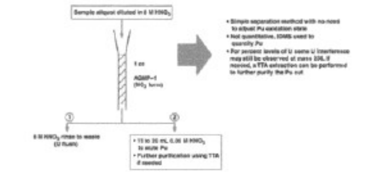
Plutonium Separations

Initial RMAL Pu Separation (7.5, 22, and 30 GWd/MT)



Procedure is time-consuming, but baseline separation of the Pu isotopes from percent levels of U isotopes is achieved

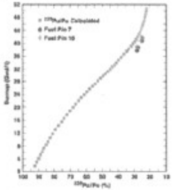
New RMAL Pu Separation (40 GWd/MT)



Comparison of ^{239}Pu/Pu quantified using the new RMAL separation and the predicted MCWO-MS method values

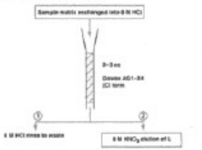
Gary Chang of INEEL has developed a method to calculate burnup based on the ratio of ^{239}Pu to total Pu.* RMAL's measured results using the new separation procedure and IDMS compare closely to the MCWO calculated values

*Chang, C. S., "ATR WG-MOX Fuel Pellet Burnup Measurement by Monte Carlo - Mass Spectrometric Method," Physor 2002, Seoul, Korea, October 7-10, 2002



Uranium Separations

RMAL U Separation (7.5, 22, and 30 GWd/MT)



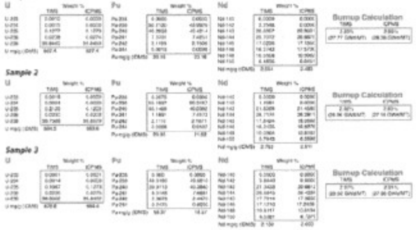
- Observed natural U contamination on resin resulting in extra time and effort spent cleaning resin prior to separations
- Proposed that a U/Pu separation was not necessary due to the low mass levels of ^{238}Pu in fuel. ^{238}Pu measured at less than 1.0% by weight while ^{238}U was greater than 99%
- These 30 GWd samples were analyzed for U isotopics with and without a separation

Pellet	^{238}U measured weight percent	
	Separation	No Separation
1	[illegible]	[illegible]
2	[illegible]	[illegible]
3	[illegible]	[illegible]

- Quantified U isotopes with no separation for the 40 GWd/MT tests

Data Comparisons

Comparison of TIMS and ICPMS Burnup Data (30 GWd/MT)



Comparisons of ICPMS Measured Burnups to Program-Calculated Burnups (40 GWd/MT)