



TRW Nuclear Energy

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**TRW Yucca Mountain Project  
Test Report  
Phase I**

**Ref. TRW Purchase Order  
No. A09112CC8A**

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## Foreward

The Waste Package Development Department of the Yucca Mountain Project has a requirement for assay data on typical commercial spent fuel to validate disposal criticality analysis methodology. Spent fuel samples from fuel rods available at General Electric's Vallecitos Nuclear Center were selected for this program. The Data generated by this program are extremely important for developing defensible criticality analysis in support of the license application for this disposal of commercial spent fuel in the Yucca Mountain repository.

This report summarizes the results for Phase I of the 2-Phase program.

## 1. Introduction

Eight samples of irradiated fuel were dissolved in the Hot Cells of GE's Vallecitos Nuclear Center (VNC) and then analyzed for selected isotopes of U, Pu, Nd, Gd, Sm, Eu, Am, Cm, Np, and Cs. Isotope concentrations, relative to U-238, were determined primarily using isotope-dilution mass spectrometry (IDMS). (A few of the isotopes were analyzed using alpha spectrometry or gamma spectrometry). Table 1-1 describes details relating to each of the 8 samples.

## 2. Experimental Procedure

### 2.1 Sample Dissolution

In the VNC Hot Cells, each sample (½-inch long, with cladding intact) was placed into a dissolver solution consisting of 100 mls of 12 M HNO<sub>3</sub> + 0.05 M HF. Sample dissolution was then allowed to proceed, at ambient temperature, over the next 4-5 days. Each sample solution was mixed several times during the dissolution period.

At the end of the dissolution period, each sample was mixed one final time. A small aliquot (~1 ml) of each solution was subsequently transferred to the Radiochemistry Laboratory for dilution and analysis.

**Table 1-1 Description of Fuel Samples**

TRW Sample ID	Fuel Rod Identification	Sub-Sample	In-Reactor Cycles	Initial Wt% U-235	Wt% Gd <sub>2</sub> O <sub>3</sub>	Sample Location*
1	TMI O1	1A2	10	4.67	0	15.5-16.0
3	TMI O1	4C2D2	10	4.67	0	109.5-110.0
4	TMI O12	1A2	10	4.67	0	15.5-16.0
6	TMI O12	4A3C2	10	4.67	0	109.5-110.0
11	QC-1 B1	3B	5-9, 11-12	3.8	0	91.5-92.0
12	QC-1 B1	5A2	5-9, 11-12	3.8	0	130.0-130.5
13	QC-1 C7	5A2	6-9, 11-12	3.0	0	130.0-130.5
14	QC-1 G5	5A2	5-9, 11-12	3.0	2.0	130.0-130.5

\*Inches above bottom end plug tip

### 2.2 Preparation of a Working Solution

A portion of each sample received by the Radiochemistry Laboratory was diluted to a fuel concentration of approximately 2.5 mg of U-238 per milliliter of solution. The diluent was 7 M nitric acid. The volume of "working solution" was about 19 milliliters. Working solutions were stored in 20 ml glass vials with "poly-seal" caps to minimize changes in concentration due to evaporation.

### 2.3 "Spike" Isotope Preparation and Calibration

Isotope "spikes", purchased from Oak Ridge National Laboratory, were prepared for use in the analysis of Gd, Sm, and Eu by isotope-dilution mass spectrometry. (Calibrated "spikes" for U, Pu, and Nd were already available).

Several milligrams of each new "spike" isotope, as the oxide, were weighed out and then dissolved in high-purity nitric acid. The dissolved "spikes" were then diluted to 100 ml.

Calibration of the new "spikes" was done by spiking a known volume of NIST-traceable elemental standards (Gd, Sm, and Eu) with a known volume of the new "spike" solutions. A thermal-ionization mass spectrometer was used to precisely measure the resulting isotope ratios. (The isotope ratios were then converted into concentration values for the "spike" solutions).

Where isotope "spike" concentrations were found to be too high for accurate sample spiking (Sm-152 and Gd-158), 1:25 ml dilutions were made prior to use with fuel samples.

### 2.4 Analysis of Isotopes of Uranium

An aliquot of each sample working solution was spiked with 0.986 ml of a burnup "triple-spike" ("0-3-2"). This "triple-spike" contains  $2.726 \times 10^{16}$  atoms of U-233 per milliliter of solution (along with Pu-242 and Nd-150).

The spike + sample mixtures were subsequently separated by ion exchange into pure samples of uranium suitable for mass spectrometric analysis. Precise isotope ratios for U-238, U-236, U-235, and U-234, relative to U-233, were determined. From the U-238/U-233 ratio, the concentration of U-238 (mg/ml) was determined. The other three uranium isotopes were then calculated relative to U-238 and were reported as mg per mg U-238.

### 2.5 Analysis of Isotopes of Plutonium

Using the same sample and spike solution from the uranium determination, spiked plutonium was isolated (after oxidizing and reducing each sample to insure that the plutonium from the sample and spike were in an identical oxidation state.)

Ion exchange procedures were used to produce pure samples of plutonium suitable for mass spectrometric analysis. Precise isotope ratios for Pu-239, Pu-240, and Pu-241, relative to Pu-242, were determined.

Since Pu-242 was present in both the sample and the "spike", a non-spiked plutonium (called Pu-unspike) sample was separated from a second portion of working solution. This second sample was processed for plutonium (and neodymium) in an as-received condition, without any "spike" additions. The Pu-unspike samples were analyzed on the mass spectrometer, measuring isotope ratios for Pu-239, Pu-240, and Pu-241, relative to Pu-242.



From the isotope ratios of the spiked and unspiked plutonium samples, concentrations of the isotopes were calculated.

## 2.6 Analysis of Pu-238

Due to the potential for U-238 interference in the mass spectrometer measurement of Pu-238 (either from residual traces of sample U-238 or from U-238 "background" in the mass spectrometer itself), alpha spectrometry was used to measure the concentration of Pu-238.

For the determination of Pu-238, a small amount of the Pu-unspike material, purified for mass spectrometric analysis, was evaporated onto a stainless steel counting disk. Each disk was then analyzed using an alpha spectrometer, resulting in a spectrum of alpha counts. (The Pu-238 atoms produce alpha-particles at 5.5 MeV, with Pu-239 and Pu-240 both producing alpha-particles at 5.15 MeV).

By combining the isotopic abundances of the Pu-239 and Pu-240 from the mass spectrometric Pu-unspike analysis with the alpha spectrometry activity ratio of  $\text{Pu-238}\alpha / (\text{Pu-239}\alpha + \text{Pu-240}\alpha)$ , the isotopic abundance of Pu-238 was determined. Concentrations are reported as mg Pu-238 per mg U-238.

## 2.7 Analysis of isotopes of Neodymium

Using the same sample + spike solution from the uranium determination, spiked neodymium was recovered. As before, ion-exchange procedures were used to isolate pure samples of neodymium suitable for mass spectrometric analysis. Precise isotope ratios for Nd-143, Nd-145, Nd-146, and Nd-148, relative to Nd-150, were determined.

Since Nd-150 was present in both the sample and the "spike", a non-spiked neodymium sample was separated from a second portion of working solution (the same non-spiked sample was also used for Pu-unspike). The purified Nd-unspike samples were also analyzed by mass spectrometry.

From the isotope ratios determined for the spiked and unspiked neodymium samples, Nd isotope concentrations were calculated.

## 2.8 Analysis of Cs-134 and Cs-137

A 25 microliter portion of each working solution was pipetted onto an absorbent 1-inch counting disk. Using gamma spectrometry, the Cs-134 content of each disk was determined from the gamma count-rate at 604.7 and 795.8 KeV, while the Cs-137 content was determined at 661.7 KeV. From the activity rates, mg Cs-134/ml and mg Cs-137/ml were calculated. Finally, the uranium concentrations (determined earlier by IDMS) were used to calculate mg Cs-134/mg U-238 and mg Cs-137/mg U-238. The results are as the date of analysis.

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## 2.9 Analysis of Isotopes of Samarium

A precisely measured volume of working solution was taken from each of the eight samples and "triple-spiked" with precise volumes of calibrated Sm-152 (diluted), Eu-153 and Gd-158 (diluted) "spikes". Two preliminary ion-exchange columns removed most of the uranium and plutonium from the samples. Finally each sample was passed through a 12-cm long column of nitrate-form AGMP-1 macroporous resin. The eluant for separating the three rare-earths (Gd, Eu, Sm) was a methanol solution containing 10% 1.6 molar HNO<sub>3</sub> (in water). A gradient was established whereby the acid concentration dropped by 12% each time 3 mls of eluant was dispensed to the column (see Figure 2-1). As shown in Figure 2-2, this allowed Gd, Eu, and Sm to be separated over a period of about 5 hours on the column. (A check on the elution of Curium on this column showed that it elutes in the same location found for Sm).

In addition to the spiked samarium, unspiked samarium samples were also required. Although a separate sample could have been taken from each working solution, it was decided to take unspiked samarium samples from previously purified curium samples, since they (Sm and Cm) elute simultaneously from the 12-cm AGMP-1 column. Very small Sm amounts (0.5 to 5 nanograms) were taken for mass spectrometric analysis to limit the alpha hazard from the curium.

From the isotope ratios determined for the spiked and unspiked samarium samples, concentrations of the isotopes were calculated (relative to U-238).

## 2.10 Analysis of isotopes of Europium

Europium from the same "triple-spiked" sample used for samarium was collected from the 12-cm AGMP-1 column as shown in Figure 2-2. A small amount of the purified Eu (~10 nanograms Eu) was subsequently measured by mass spectrometry.

In addition to the spiked europium, unspiked europium was separated from a second aliquot of working solution (without added spikes). Again, a 12-cm AGMP-1 column was used to purify the Eu. A small portion of the purified Eu (~10 nanograms) was then analyzed by mass spectrometry.

Based on the results from the mass spectrometric analysis of the spiked and unspiked Eu, an improved spiking procedure was implemented. Instead of using the Eu results from Eu-153 spiking, new samples were pipetted from the working solutions and spiked with about half the amount of the "alternative" spike, Eu-151. This change resulted in ideal isotope ratios and greatly improved the accuracy of the analysis. After purification on 12-cm AGMP-1 columns the respiked Eu samples were analyzed by mass spectrometry.

From the isotope ratios determined for the (Eu-151) spiked and unspiked europium samples, concentrations of the isotopes, relative to U-238, were calculated.

## 2.11 Analysis of Gd-155

Gadolinium from each "triple-spiked" sample, except #14, was collected from the 12-cm AGMP-1 column as shown in Figure 2-2. A small amount of the purified Gd (~40 ng) was subsequently measured by mass spectrometry.

**Note:** Sample #14, which was manufactured with 2 wt% Gd<sub>2</sub>O<sub>3</sub>, was spiked separately using 0.100 ml of working solution and 1.0 ml of "concentrated" Gd-158 "spike". The "concentrated spike" contained 25 times more Gd-158 per ml than did the "diluted spike".

Unspiked gadolinium from a second aliquot of working solution was also separated on a 12-cm AGMP-1 column.

Mass spectrometry showed the presence of an interfering mass in the gadolinium samples, especially the unspiked Gd at mass 155. The spiked Gd samples also showed the interference at mass 155, but showed no affect on the Gd-156/Gd-158 ratio, which is the only ratio required from the spiked gadolinium samples.

In order to improve the Gd-155 measurement, new working solution samples were taken and the gadolinium separated on 12-cm AGMP-1 columns. The gadolinium fraction was then repurified on a second column, 12-cm long, of AGMP-1 resin to produce higher purity gadolinium. Finally, these double-purified Gd samples were fumed with nitric acid plus a drop of perchloric acid. Again, ~40 nanogram samples of Gd were analyzed by mass spectrometry.

The repurified gadolinium samples showed decreased interference when analyzed on the mass spectrometer. All samples did, however, continue to show some residual mass 155 interference, which decreased with time. After running times of 3 to 4 hours, a sample would typically show a 155/158 ratio changing less than 1% over 10 minutes. Calculations of Gd-155 concentrations were based on the final measurement series, representing the last 10 minutes of analysis.

## 2.12 Analysis of Isotopes of Curium

For curium isotopic analysis, a 50 microliter sample from each working solution was diluted to 10.0 ml. From this dilution an exact volume (25 or 50 microliters) was dried onto the surface of a stainless steel counting planchet. The alpha planchets were then alpha counted using a gas proportional alpha counter with known efficiency. The same planchets were then analyzed by alpha spectrometry to determine the fraction of alpha counts due to Cm-242 (6.1 MeV) as well as the sum of Cm-243 plus Cm-244 (5.8 MeV).

Larger samples of working solution (1 ml from QC-1 samples, 2-3 ml from TMI samples) were then taken for isolating Cm and Am for mass spectrometric measurements. These samples were prepurified on ion exchange columns of Dowex 1-X4 resin to remove most of the uranium and plutonium before proceeding with the chromatographic separation of Am and Cm.

Americium and curium were isolated using 12-cm long columns of nitrate-form AGMP-1 macroporous resin. The eluant for separating Am and Cm was a methanol solution containing 10% 0.3 M HNO<sub>3</sub> (in water). During elution, a gradient was established whereby the acid concentration dropped by 12% each time 3 ml of eluant was dispensed to the column. Figure 2-3 shows the change in acid normality over the course of the separation process. The elution of Cm and Am under these conditions is shown in Figure 2-4.

A small amount of the purified Cm (0.3-0.5 nanograms) was subsequently deposited on a rhenium ribbon filament and analyzed for relative abundances of the curium isotopes by thermal ionization mass spectrometry. From the alpha analysis (combined with the mass spectrometric analysis), the concentration of each Cm isotope relative to U-238 was calculated. Cm-242 was reported both from direct alpha spectrometry analysis and from mass spectrometric determination of the Cm-242/Cm-244 ratio.

### 2.13 Analysis of Isotopes of Americium

Americium was benchmarked to U-238 (through the curium concentration) by analyzing a portion of the working solution after it had passed through a nitrate-form Dowex 1-X4 column. (This nitrate form column removed plutonium, but allowed Am and Cm to pass through unaffected by the column). Once the plutonium had been removed, a small portion (50 microliters) of the resultant solution was evaporated onto a stainless steel planchet. The resultant alpha spectrum, free of interfering Pu-238, allowed Am-241 (5.5 MeV) to be related to the sum peak of Cm-243 plus Cm-244 (5.8 MeV).

After eluting the Am fraction from the 12-cm AGMP-1 column (detailed in the curium section), a small amount of the purified Am (3-6 nanograms) was subsequently deposited on a rhenium ribbon filament and analyzed for relative abundances of the americium isotopes by thermal ionization mass spectrometry.

From the alpha analysis (combined with the mass spectrometric analysis), the concentration of the Am isotopes relative to U-238 was calculated.

### 2.14 Analysis of Np-237

A two milliliter sample of each working solution was spiked with a known quantity of Np-239 tracer ( $t_{1/2} = 2.35$  days). Oxidation states were reduced using an excess of ferrous ammonium sulfate. After 30 minutes, each sample solution was passed through a 2 cm long column of nitrate-form AG1-X4 ion exchange resin. The Np was absorbed by the resin, while fission products, uranium and plutonium were washed off with 7 M nitric acid followed by 0.1 M NH<sub>4</sub>I in 12 M HCL. The Np was then eluted from the column and transferred to a stainless steel alpha planchet.

Each alpha planchet was counted by gamma spectrometry to determine the chemical recovery of the added Np-239 "spike". Then the planchets were counted for total alpha count-rate using a calibrated proportional counter. Finally, each planchet was analyzed by alpha spectroscopy to determine the fraction of alpha counts due to Np-237 (4.8 MeV).

Since "excess" Np-239, above the amount added, (produced from the alpha decay of Am-243,  $t_{1/2} = 7370$  years) was found to be present in all samples a correction to the observed Np-239 yield was made based on the analysis of Am-243 in each sample. Since the QC-1 samples contained the most Am-243, the correction for those 4 samples was large. To improve the accuracy of the method, the four QC-1 samples were resampled from the working solutions and respiked with 10 times more Np-239 tracer.

The four respiked Np-237 samples from QC-1 were purified by ion exchange and analyzed as described above. The resultant Np-237 concentrations were recalculated as before, requiring less correction for Np-239 from Am-243 decay. All four respiked samples gave Np-237 concentrations within  $\pm 2.7\%$  (relative) of the concentration determined from the original analysis.

## 2.15 Burnup Determinations

Burnup was calculated for all samples using GE's BRNUP (VAX) code. Burnup was calculated using both our "normal" inputs (U-spiked, Nd-spiked, Pu-spiked and Pu-unspiked) and the addition of the unspiked neodymium mass ratios. (The burnup code is designed to calculate burnup with or without unspiked neodymium values).

Figure 2-1.  
Nitric acid "Gradient" concentration  
(For Rare-earth separations)

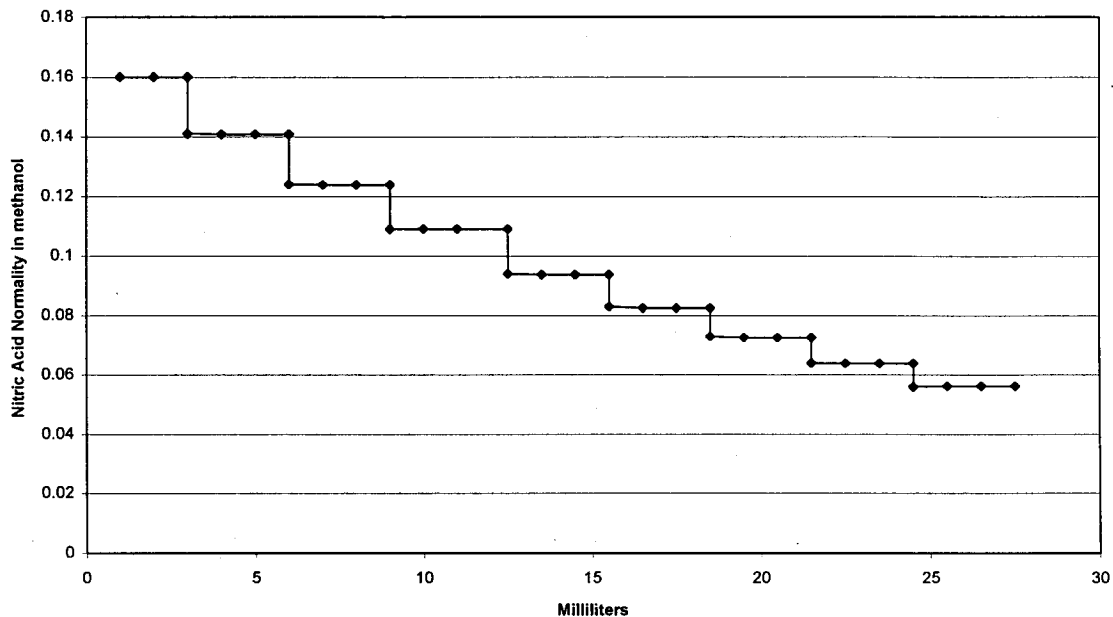


Figure 2-2.  
Rare-Earth Separations on 12-cm AGMP-1 nitrated resin  
Gradient, starting with 1.6 Molar Nitric acid (10% in methanol)

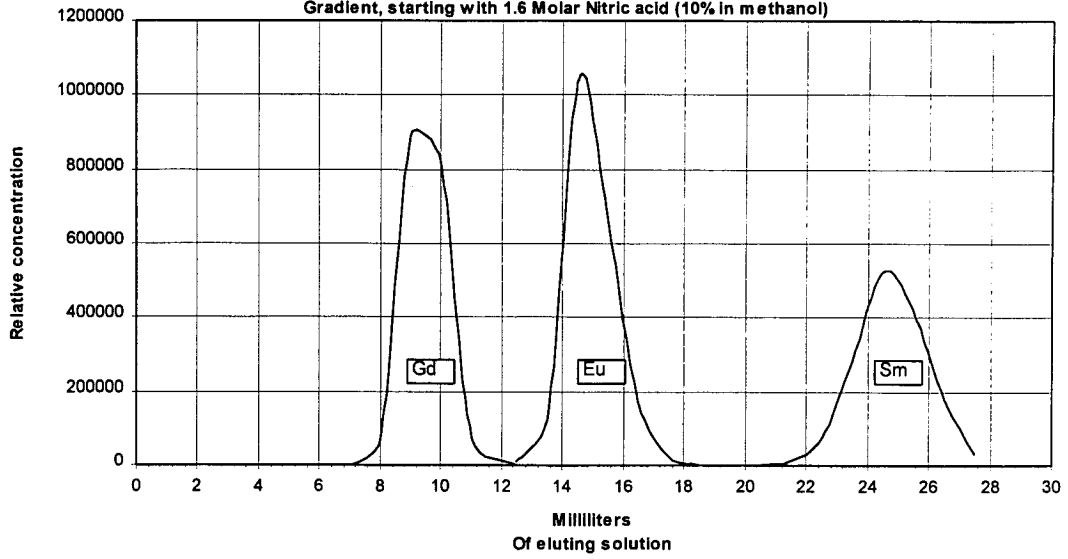


Figure 2-3.  
Nitric acid "Gradient" concentration  
(For Am-Cm separations)

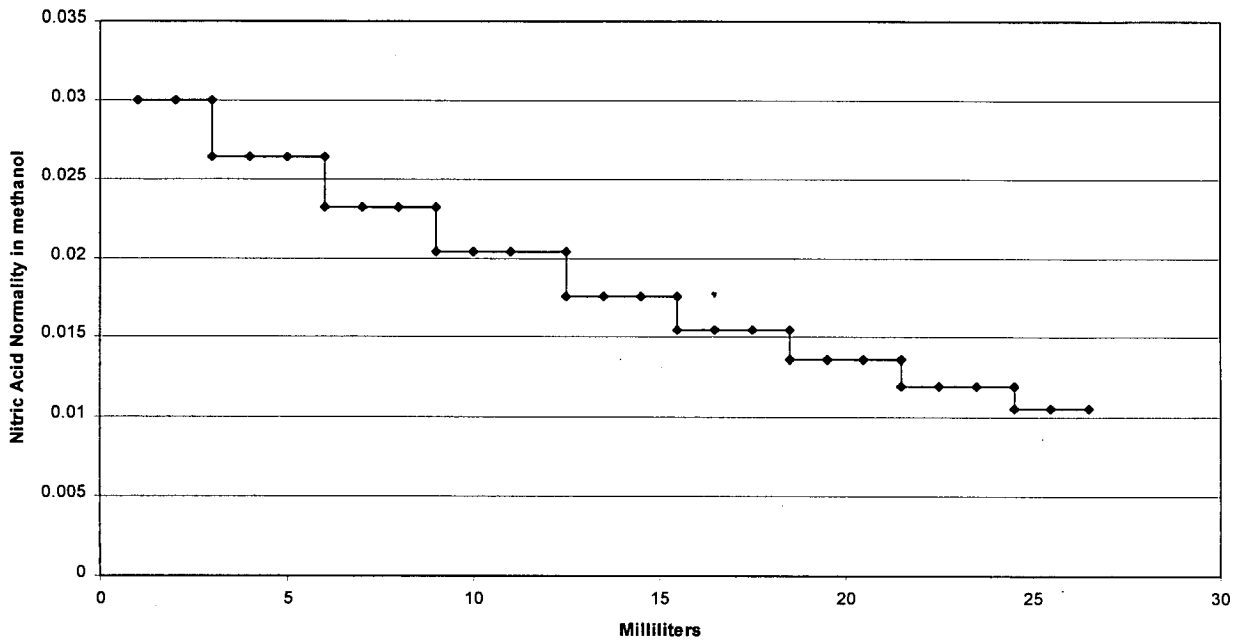
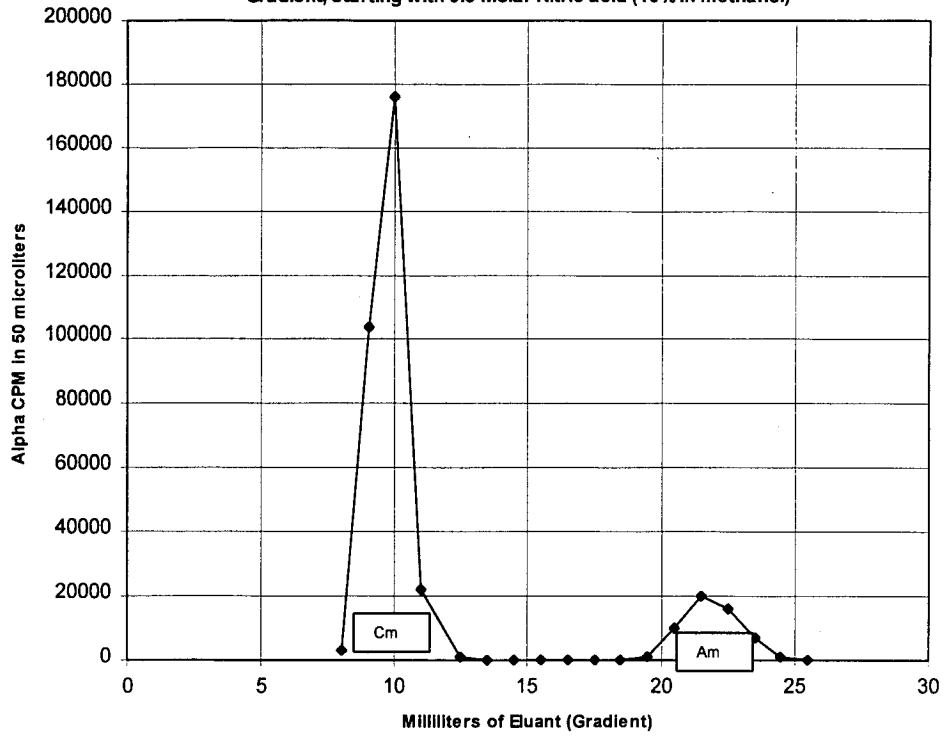


Figure 2-4.

Am and Cm Elution on 12-cm AGMP-1 nitrated resin  
Gradient, starting with 0.3 Molar Nitric acid (10% in methanol)



### 3. Results

The isotopic concentrations and burnup results for the 8 fuel samples investigated under Phase I of this contract are shown in Tables 3-1 through 3-11. Table 3-12 summarizes the estimated relative uncertainty, at the 95% confidence level, of each measurement.

**Table 3-1  
Burnup Analysis Summary**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub-sample</u>	<u>"Normal" Nd Burnup Anal.</u>	<u>Including "non-spiked" Nd analysis</u>	<u>Burnup average MWd/MTU</u>
1	TMI O1	1A2	2.58E+04	2.58E+04	2.58E+04
3	TMI O1	4C2D2	2.66E+04	2.67E+04	2.67E+04
4	TMI O12	1A2	2.37E+04	2.37E+04	2.37E+04
6	TMI O12	4A3C2	2.40E+04	2.40E+04	2.40E+04
11	QC-1 B1	3B	7.32E+04	7.59E+04	7.46E+04
12	QC-1 B1	5A2	5.31E+04	5.31E+04	5.31E+04
13	QC-1 C7	5A2	5.52E+04	5.49E+04	5.50E+04
14	QC-1 G5	5A2	4.56E+04	4.53E+04	4.55E+04

**Table 3-2  
Uranium Isotope Concentrations**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub-sample</u>	<u>mg U-234 per mg U-238</u>	<u>mg U-235 per mg U-238</u>	<u>mg U-236 per mg U-238</u>
1	TMI O1	1A2	3.48E-04	2.35E-02	4.83E-03
3	TMI O1	4C2D2	3.35E-04	2.32E-02	4.99E-03
4	TMI O12	1A2	3.55E-04	2.51E-02	4.58E-03
6	TMI O12	4A3C2	3.48E-04	2.55E-02	4.68E-03
11	QC-1 B1	3B	1.54E-04	1.12E-03	5.93E-03
12	QC-1 B1	5A2	1.82E-04	3.75E-03	5.97E-03
13	QC-1 C7	5A2	1.28E-04	1.39E-03	4.62E-03
14	QC-1 G5	5A2	1.60E-04	3.30E-03	4.56E-03



**Table 3-3  
Plutonium Isotope Concentrations**

<u>TRW</u> <u>Sample ID</u>	<u>Fuel Rod</u> <u>Identification</u>	<u>Sub-</u> <u>sample</u>	<u>Mg Pu238</u> <u>Per mg U238</u>	<u>mg Pu-239</u> <u>per mg U-238</u>	<u>mg Pu-240</u> <u>per mg U-238</u>	<u>mg Pu-241*</u> <u>per mg U-238</u>	<u>mg Pu-242</u> <u>per mg U-238</u>
1	TMI O1	1A2	7.67E-05	5.81E-03	1.62E-03	8.04E-04	1.92E-04
3	TMI O1	4C2D2	1.00E-04	6.44E-03	1.83E-03	9.56E-04	2.36E-04
4	TMI O12	1A2	6.68E-05	5.79E-03	1.48E-03	7.34E-04	1.58E-04
6	TMI O12	4A3C2	8.29E-05	6.60E-03	1.61E-03	8.54E-04	1.76E-04
11	QC-1 B1	3B	6.58E-04	4.04E-03	3.61E-03	1.04E-03	1.99E-03
12	QC-1 B1	5A2	5.41E-04	4.61E-03	3.38E-03	1.08E-03	1.23E-03
13	QC-1 C7	5A2	4.58E-04	3.92E-03	3.30E-03	9.49E-04	1.47E-03
14	QC-1 G5	5A2	4.64E-04	5.80E-03	3.06E-03	1.16E-03	9.87E-04

\* As of 4/7/99

**Table 3-4  
Neodymium Isotope Concentrations**

<u>TRW</u> <u>Sample ID</u>	<u>Fuel Rod</u> <u>Identification</u>	<u>Sub-</u> <u>sample</u>	<u>mg Nd-143</u> <u>per mg U-238</u>	<u>mg Nd-145</u> <u>per mg U-238</u>	<u>Mg Nd-146</u> <u>Per mg U-238</u>	<u>mg Nd-148</u> <u>per mg U-238</u>	<u>mg Nd-150</u> <u>per mg U-238</u>
1	TMI O1	1A2	7.95E-04	6.00E-04	5.56E-04	3.05E-04	1.38E-04
3	TMI O1	4C2D2	8.28E-04	6.21E-04	5.87E-04	3.21E-04	1.47E-04
4	TMI O12	1A2	7.51E-04	5.59E-04	5.12E-04	2.81E-04	1.26E-04
6	TMI O12	4A3C2	7.66E-04	5.64E-04	5.26E-04	2.88E-04	1.31E-04
11	QC-1 B1	3B	8.77E-04	1.23E-03	1.70E-03	8.02E-04	4.13E-04
12	QC-1 B1	5A2	9.75E-04	1.04E-03	1.25E-03	6.22E-04	3.11E-04
13	QC-1 C7	5A2	7.60E-04	9.70E-04	1.23E-03	6.05E-04	3.11E-04
14	QC-1 G5	5A2	8.58E-04	8.75E-04	1.04E-03	5.24E-04	2.67E-04

**Table 3-5  
Cesium Isotope Concentrations**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub-sample</u>	<u>mg Cs-134 per mg of U-238 at count-time*</u>	<u>mg Cs-137 per mg of U-238 at count-time*</u>
1	TMI O1	1A2	2.51E-05	9.71E-04
3	TMI O1	4C2D2	2.90E-05	1.03E-03
4	TMI O12	1A2	2.22E-05	9.05E-04
6	TMI O12	4A3C2	2.44E-05	9.18E-04
11	QC-1 B1	3B	2.49E-05	2.08E-03
12	QC-1 B1	5A2	1.77E-05	1.63E-03
13	QC-1 C7	5A2	1.77E-05	1.58E-03
14	QC-1 G5	5A2	1.28E-05	1.36E-03

\* As of 3/19/99

**Table 3-6  
Europium Isotope Concentrations**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub-sample</u>	<u>mg Eu-151 per mg U-238</u>	<u>mg Eu-153 per mg U-238</u>
1	TMI O1	1A2	4.15E-07	8.05E-05
3	TMI O1	4C2D2	4.61E-07	8.80E-05
4	TMI O12	1A2	4.29E-07	7.37E-05
6	TMI O12	4A3C2	4.89E-07	7.69E-05
11	QC-1 B1	3B	5.57E-07	2.35E-04
12	QC-1 B1	5A2	5.53E-07	1.93E-04
13	QC-1 C7	5A2	4.67E-07	1.89E-04
14	QC-1 G5	5A2	6.71E-07	1.73E-04

**Table 3-7  
Samarium Isotope Concentrations**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub- sample</u>	<u>mg Sm-147 per mg U-238</u>	<u>mg Sm-149 per mg U-238</u>	<u>mg Sm-150 per mg U-238</u>	<u>mg Sm-151 per mg U-238</u>	<u>mg Sm-152 per mg U-238</u>
1	TMI O1	1A2	1.91E-04	4.32E-06	2.30E-04	1.36E-05	9.23E-05
3	TMI O1	4C2D2	1.94E-04	4.72E-06	2.47E-04	1.53E-05	9.54E-05
4	TMI O12	1A2	1.81E-04	4.32E-06	2.11E-04	1.38E-05	8.62E-05
6	TMI O12	4A3C2	1.79E-04	4.73E-06	2.17E-04	1.58E-05	8.41E-05
11	QC-1 B1	3B	3.84E-04	1.62E-06	5.28E-04	1.05E-05	1.88E-04
12	QC-1 B1	5A2	3.81E-04	1.97E-06	4.28E-04	1.08E-05	1.57E-04
13	QC-1 C7	5A2	3.42E-04	1.55E-06	4.06E-04	8.96E-06	1.58E-04
14	QC-1 G5	5A2	3.25E-04	1.97E-06	3.69E-04	1.24E-05	1.25E-04

**Table 3-8  
Gd-155 Concentrations**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub- sample</u>	<u>mg Gd-155 per mg U-238</u>
1	TMI O1	1A2	2.46E-06
3	TMI O1	4C2D2	2.82E-06
4	TMI O12	1A2	2.03E-06
6	TMI O12	4A3C2	2.33E-06
11	QC-1 B1	3B	1.30E-05
12	QC-1 B1	5A2	9.99E-06
13	QC-1 C7	5A2	1.02E-05
14	QC-1 G5	5A2	1.23E-05

**Table 3-9  
Curium Isotope Concentrations**

TRW Sample ID	Fuel Rod Identification	Sub- sample	(Alpha Spec)	(Mass Spec)	mg Cm243 per mg U238	mg Cm244 per mg U238	mg Cm245 per mg U238
			mg Cm242* per mg U238	mg Cm242* per mg U238			
1	TMI O1	1A2	1.5E-08	2.2E-08	5.50E-08	2.66E-06	1.19E-07
3	TMI O1	4C2D2	2.6E-08	3.2E-08	1.04E-07	5.32E-06	2.81E-07
4	TMI O12	1A2	1.9E-08	2.7E-08	6.36E-08	2.89E-06	1.24E-07
6	TMI O12	4A3C2	1.8E-08	2.2E-08	6.99E-08	3.22E-06	1.67E-07
11	QC-1 B1	3B	ND	ND	1.82E-06	3.00E-04	1.91E-05
12	QC-1 B1	5A2	ND	ND	1.46E-06	1.12E-04	7.83E-06
13	QC-1 C7	5A2	ND	ND	1.31E-06	1.48E-04	8.75E-06
14	QC-1 G5	5A2	ND	ND	1.09E-06	1.01E-04	9.44E-06

\* As of 3/29/99

ND =Not Detected

**Table 3-10  
Americium Isotope Concentrations**

TRW Sample ID	Fuel Rod Identification	Sub- sample	mg Am241 per mg U238	mg Am242m per mg U238	mg Am243 per mg U238
1	TMI O1	1A2	1.22E-04	2.93E-07	1.60E-05
3	TMI O1	4C2D2	1.83E-04	4.50E-07	2.74E-05
4	TMI O12	1A2	1.62E-04	3.77E-07	1.80E-05
6	TMI O12	4A3C2	1.47E-04	3.97E-07	1.76E-05
11	QC-1 B1	3B	5.02E-04	1.53E-06	5.64E-04
12	QC-1 B1	5A2	5.14E-04	2.21E-06	2.89E-04
13	QC-1 C7	5A2	4.38E-04	1.37E-06	3.55E-04
14	QC-1 G5	5A2	5.64E-04	3.65E-06	2.48E-04

**Table 3-11  
Np-237 Concentrations**

<u>TRW</u> <u>Sample ID</u>	<u>Fuel Rod</u> <u>Identification</u>	<u>Sub-</u> <u>sample</u>	<u>mg Np237</u> <u>per mg U238</u>	<u>Re-spiked</u> <u>Replicate</u>	<u>Average</u> <u>mg Np237</u> <u>per mg U238</u>
1	TMI O1	1A2	3.24E-04		
3	TMI O1	4C2D2	3.89E-04		
4	TMI O12	1A2	3.23E-04		
6	TMI O12	4A3C2	3.50E-04		
11	QC-1 B1	3B	7.95E-04	7.79E-04	7.87E-04
12	QC-1 B1	5A2	6.60E-04	6.78E-04	6.69E-04
13	QC-1 C7	5A2	5.75E-04	5.64E-04	5.70E-04
14	QC-1 G5	5A2	5.64E-04	5.79E-04	5.71E-04

**Table 3-12  
Estimated Measurement Uncertainties**

Isotope*	Estimated percentage uncertainty @95% confidence
U-238	1
U-234	1
U-235	1
U-236	1
Cs-134	3.5
Cs-137	3.5
Pu-238	5
Pu-239	1.2
Pu-240	1.2
Pu-241	1.2
Pu-242	1.2
Nd-143	1.5
Nd-145	1.5
Nd-146	1.5
Nd-148	1.5
Nd-150	1.5
Sm-147	1.7
Sm-149	1.8
Sm-150	1.7
Sm-151	1.7
Sm-152	1.7
Eu-151	1.7
Eu-153	1.8
Gd-155	2.7**
Am-241	7
Am-242	7
Am-243	7
Cm-242	20
Cm-243	5.5
Cm-244	5.5
Cm-245	5.5
Np-237	5.8

\*reported as mg/mg U238 (except for the U-238 measurement)

\*\*Interference

#### 4.0 Reference Materials

1. Burnup "Triple-spike" - - "0-3-2"  
Contains: 2.726x10<sup>16</sup> Atoms U-233/ml  
3.489x10<sup>16</sup> Atoms Pu-242/ml  
1.817x10<sup>15</sup> Atoms Nd-150/ml
2. Gd-158 - - ORNL Batch 109690  
97.5% Isotopically Pure
3. Sm-152 - - ORNL Batch 167692  
99.4% Isotopically Pure
4. Eu-151 - - ORNL Batch 151892  
96.8% Isotopically Pure
5. Gadolinium, Standard, 10,000 µg/ml  
Traceable to NIST SRM 928  
Lot No. ID-0887  
ULTRA Scientific
6. Samarium Standard, 10,000 µg/ml  
Traceable to NIST SRM 3147  
Lot No. ID-0641  
ULTRA Scientific
7. Europium Standard, 10,000 µg/ml  
Traceable to NIST SRM 3117  
Lot No. ID-0637  
ULTRA Scientific
8. Plutonium Isotopic Standard, SRM 946  
NIST
9. Uranium Isotopic Standards, U010 and U050  
NIST

**5. Equipment List**

1. Thermal Ionization Mass Spectrometer

Finnigan MAT  
Model 261

2. Gamma Spectrometer

Nuclear Data 6700  
Ortec HPGe detector

3. Proportional Counter

NMC  
Model PC-5

4. Alpha Spectrometer

Ortec  
Model 676A  
Plus GENIE 2000 Analyzer (Canberra)



**TRW Yucca Mountain Project  
Test Report  
Phase 2**

**Ref. TRW Purchase Order  
No. A09112CC8A**



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*General Electric*

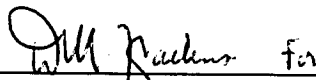
**TRW Yucca Mountain Project  
Test Report  
Phase 2**

**Ref. TRW Purchase Order  
No. A09112CC8A**



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R. D. Reager  
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Materials Technology

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## Foreward

The Waste Package Development Department of the Yucca Mountain Project has a requirement for assay data on typical commercial spent fuel to validate disposal criticality analysis methodology. Spent fuel samples from fuel rods available at General Electric's Vallecitos Nuclear Center were selected for this program. The Data generated by this program are extremely important for developing defensible criticality analysis in support of the license application for the disposal of commercial spent fuel in the Yucca Mountain repository.

This report summarizes the results for Phase 2 of the 2-Phase program.

## 1. Introduction

Six samples of irradiated fuel were dissolved in the Hot Cells of GE's Vallecitos Nuclear Center (VNC) and then analyzed for selected isotopes of U, Pu, Nd, Gd, Sm, Eu, Am, Cm, Np, and Cs. Isotope concentrations, relative to U-238, were determined primarily using isotope-dilution mass spectrometry (IDMS). (A few of the isotopes were analyzed using alpha spectrometry or gamma spectrometry). Table 1-1 describes details relating to each of the 6 samples.

## 2. Experimental Procedure

### 2.1 Sample Dissolution

In the VNC Hot Cells, each sample (1/2-inch long, with cladding intact) was placed into a dissolver solution consisting of 100 ml of 12 M HNO<sub>3</sub> + 0.05 M HF. Sample dissolution was then allowed to proceed, at ambient temperature, over the next 4-5 days. Each sample solution was mixed several times during the dissolution period.

At the end of the dissolution period, each sample was mixed one final time. A small aliquot (~1 ml) of each solution was subsequently transferred to the Radiochemistry Laboratory for dilution and analysis.

**Table 1-1 Description of Fuel Samples**

TRW Sample ID	Fuel Rod Identification	Sub-Sample	In-Reactor Cycles	Initial Wt% U-235	Wt% Gd <sub>2</sub> O <sub>3</sub>	Sample Location*
2	TMI O1	3A2	10	4.67	0	77.6-78.1
5	TMI O12	3A2	10	4.67	0	77.6-78.1
7	TMI O13	1A2	10	4.67	0	15.5-16.0
8	TMI O13	3A2	10	4.67	0	77.6-78.1
9	QC-1 A2	3B1	5-9, 11-12	3.8	0	91.5-92.0
10	QC-1 A2	5A1B	5-9, 11-12	3.8	0	130.0-130.5

\*Inches above bottom end plug tip

### 2.2 Preparation of a Working Solution

A portion of each sample received by the Radiochemistry Laboratory was diluted to a fuel concentration of approximately 2.5 mg of U-238 per milliliter of solution. The diluent was 7 M nitric acid. The volume of "working solution" was about 19 milliliters. Working solutions were stored in 20 ml glass vials with "poly-seal" caps to minimize changes in concentration due to evaporation.

### 2.3 "Spike" Isotope Preparation and Calibration

Isotope "spikes", purchased from Oak Ridge National Laboratory, were prepared for use in the analysis of Gd, Sm, and Eu by isotope-dilution mass spectrometry. (Calibrated "spikes" for U, Pu, and Nd were already available).

Several milligrams of each new "spike" isotope, as the oxide, were weighed out and then dissolved in high-purity nitric acid. The dissolved "spikes" were then diluted to 100 ml.

Calibration of the new "spikes" was done by spiking a known volume of NIST-traceable elemental standards (Gd, Sm, and Eu) with a known volume of the new "spike" solutions. A thermal-ionization mass spectrometer was used to precisely measure the resulting isotope ratios. (The isotope ratios were then converted into concentration values for the "spike" solutions).

Where isotope "spike" concentrations were found to be too high for accurate sample spiking (Sm-152 and Gd-158), 1:25 ml dilutions were made prior to use with fuel samples.

### 2.4 Analysis of Isotopes of Uranium

An aliquot of each sample working solution was spiked with 0.985 ml of a burnup "triple-spike" ("0-3-2"). This "triple-spike" contains  $2.726 \times 10^{16}$  atoms of U-233 per milliliter of solution (along with Pu-242 and Nd-150).

The spike + sample mixtures were subsequently separated by ion exchange into pure samples of uranium suitable for mass spectrometric analysis. Precise isotope ratios for U-238, U-236, U-235, and U-234, relative to U-233, were determined. From the U-238/U-233 ratio, the concentration of U-238 (mg/ml) was determined. The other three uranium isotopes were then calculated relative to U-238 and were reported as mg per mg U-238.

### 2.5 Analysis of Isotopes of Plutonium

Using the same sample and spike solution from the uranium determination, spiked plutonium was isolated (after oxidizing and reducing each sample to insure that the plutonium from the sample and spike were in an identical oxidation state.)

Ion exchange procedures were used to produce pure samples of plutonium suitable for mass spectrometric analysis. Precise isotope ratios for Pu-239, Pu-240, and Pu-241, relative to Pu-242, were determined.

Since Pu-242 was present in both the sample and the "spike", a non-spiked plutonium (called Pu-unspike) sample was separated from a second portion of working solution. This second sample was processed for plutonium (and neodymium) in an as-received condition, without any "spike" additions. The Pu-unspike samples were analyzed on the mass spectrometer, measuring isotope ratios for Pu-239, Pu-240, and Pu-241, relative to Pu-242.



From the isotope ratios of the spiked and unspiked plutonium samples, concentrations of the isotopes were calculated.

## 2.6 Analysis of Pu-238

Due to the potential for U-238 interference in the mass spectrometer measurement of Pu-238 (either from residual traces of sample U-238 or from U-238 "background" in the mass spectrometer itself), alpha spectrometry was used to measure the concentration of Pu-238.

For the determination of Pu-238, a small amount of the Pu-unspike material, purified for mass spectrometric analysis, was evaporated onto a stainless steel counting disk. Each disk was then analyzed using an alpha spectrometer, resulting in a spectrum of alpha counts. (The Pu-238 atoms produce alpha-particles at 5.5 MeV, with Pu-239 and Pu-240 both producing alpha-particles at 5.15 MeV).

By combining the isotopic abundances of the Pu-239 and Pu-240 from the mass spectrometric Pu-unspike analysis with the alpha spectrometry activity ratio of  $\text{Pu-238}\alpha / (\text{Pu-239}\alpha + \text{Pu-240}\alpha)$ , the isotopic abundance of Pu-238 was determined. Concentrations are reported as mg Pu-238 per mg U-238.

## 2.7 Analysis of isotopes of Neodymium

Using the same sample + spike solution from the uranium determination, spiked neodymium was recovered. As before, ion-exchange procedures were used to isolate pure samples of neodymium suitable for mass spectrometric analysis. Precise isotope ratios for Nd-143, Nd-145, Nd-146, and Nd-148, relative to Nd-150, were determined.

Since Nd-150 was present in both the sample and the "spike", a non-spiked neodymium sample was separated from a second portion of working solution (the same non-spiked sample was also used for Pu-unspike). The purified Nd-unspike samples were also analyzed by mass spectrometry.

From the isotope ratios determined for the spiked and unspiked neodymium samples, Nd isotope concentrations were calculated.

## 2.8 Analysis of Cs-134 and Cs-137

A 25 microliter portion of each working solution was pipetted onto an absorbent 1-inch counting disk. Using gamma spectrometry, the Cs-134 content of each disk was determined from the gamma count-rate at 604.7 and 795.8 KeV, while the Cs-137 content was determined at 661.7 KeV. From the activity rates, mg Cs-134/ml and mg Cs-137/ml were calculated. Finally, the uranium concentrations (determined earlier by IDMS) were used to calculate mg Cs-134/mg U-238 and mg Cs-137/mg U-238. The results are as the date of analysis.

## 2.9 Analysis of Isotopes of Samarium

A precisely measured volume of working solution was taken from each of the six samples and "triple-spiked" with precise volumes of calibrated Sm-152 (diluted), Eu-151 and Gd-158 (diluted) "spikes". Two preliminary ion-exchange columns removed most of the uranium and plutonium from the samples. Finally each sample was passed through a 12-cm long column of nitrate-form AGMP-1 macroporous resin. The eluant for separating the three rare-earths (Gd, Eu, Sm) was a methanol solution containing 10% 1.6 molar HNO<sub>3</sub> (in water). A gradient was established whereby the acid concentration dropped by 12% each time 3 ml of eluant was dispensed to the column (see Figure 2-1). As shown in Figure 2-2, this allowed Gd, Eu, and Sm to be separated over a period of about 5 hours on the column. (A check on the elution of Curium on this column showed that it elutes in the same location found for Sm).

In addition to the spiked samarium, unspiked samarium samples were also required. Although a separate sample could have been taken from each working solution, it was decided to take unspiked samarium samples from previously purified curium samples, since they (Sm and Cm) elute simultaneously from the 12-cm AGMP-1 column. Very small Sm amounts (0.5 to 5 nanograms) were taken for mass spectrometric analysis to limit the alpha hazard from the curium.

From the isotope ratios determined for the spiked and unspiked samarium samples, concentrations of the isotopes were calculated (relative to U-238).

## 2.10 Analysis of isotopes of Europium

Europium from the same "triple-spiked" sample used for samarium was collected from the 12-cm AGMP-1 column as shown in Figure 2-2. A small amount of the purified Eu (~10 nanograms Eu) was subsequently measured by mass spectrometry.

In addition to the spiked europium, unspiked europium was separated from a second aliquot of working solution (without added spikes). Again, a 12-cm AGMP-1 column was used to purify the Eu. A small portion of the purified Eu (~10 nanograms) was then analyzed by mass spectrometry.

From the isotope ratios determined for the (Eu-151) spiked and unspiked europium samples, concentrations of the isotopes, relative to U-238, were calculated.

### 2.11 Analysis of Gd-155

Gadolinium from each "triple-spiked" sample was collected from the 12-cm AGMP-1 column as shown in Figure 2-2. A second 12-cm AGMP-1 column was then used to repurify the spiked gadolinium samples prior to mass spectrometric analysis. A small amount (~40 ng) of each doubly purified spiked Gd sample was subsequently measured by mass spectrometry.

Unspiked gadolinium was separated from a second aliquot of working solution (without added spikes). As with the spiked gadolinium, the unspiked Gd was isolated using a 12-cm AGMP-1 column. To reduce interferences to an absolute minimum the unspiked Gd samples were then repurified twice more using 12-cm AGMP-1 columns. A small amount (~40 ng) of each purified Gd sample was subsequently measured by mass spectrometry.

The mass spectrometric analyses of these highly purified gadolinium samples showed reduced interference at mass 155 when compared to the doubly-purified samples from Phase 1. Run times were in the 1-3 hour range, with precision of the 155/158 ratio better than 1% near the end of the analysis period.

From the isotope ratios determined for the spiked and unspiked gadolinium samples, concentrations of Gd-155, relative to U-238, were calculated.

### 2.12 Analysis of Isotopes of Curium

For curium isotopic analysis, a 50 microliter sample from each working solution was diluted to 10.0 ml (50 ml for samples 9 and 10). From this dilution an exact volume (25 or 50 microliters) was dried onto the surface of a stainless steel counting planchet. The alpha planchets were then alpha counted using a gas proportional alpha counter with known efficiency. The same planchets were then analyzed by alpha spectrometry to determine the fraction of alpha counts due to Cm-242 (6.1 MeV) as well as the sum of Cm-243 plus Cm-244 (5.8 MeV).

Larger samples of working solution (1 ml from QC-1 samples, 2.5 ml from TMI samples) were then taken for isolating Cm and Am for mass spectrometric measurements. These samples were prepurified on ion exchange columns of Dowex 1-X4 resin to remove most of the uranium and plutonium before proceeding with the chromatographic separation of Am and Cm.

Americium and curium were isolated using 12-cm long columns of nitrate-form AGMP-1 macroporous resin. The eluant for separating Am and Cm was a methanol solution containing 10% 0.3 M HNO<sub>3</sub> (in water). During elution, a gradient was established whereby the acid concentration dropped by 12% each time 3 ml of eluant was dispensed to the column. Figure 2-3 shows the change in acid normality over the course of the separation process. The elution of Cm and Am under these conditions is shown in Figure 2-4.

A small amount of the purified Cm (0.3-0.5 nanograms) was subsequently deposited on a rhenium ribbon filament and analyzed for relative abundances of the curium isotopes by thermal ionization mass spectrometry. From the alpha analysis (combined with the mass spectrometric analysis), the concentration of each Cm isotope relative to U-238 was calculated. Cm-242 was reported both from direct alpha spectrometry analysis and from mass spectrometric determination of the Cm-242/Cm-244 ratio.

#### 2.13 Analysis of Isotopes of Americium

Americium was benchmarked to U-238 (through the curium concentration) by analyzing a portion of the working solution after it had passed through a nitrate-form Dowex 1-X4 column. (This nitrate form column removed plutonium, but allowed Am and Cm to pass through unaffected by the column). Once the plutonium had been removed, a small portion (50 microliters) of the resultant solution was evaporated onto a stainless steel planchet. The resultant alpha spectrum, free of interfering Pu-238, allowed Am-241 (5.5 MeV) to be related to the sum peak of Cm-243 plus Cm-244 (5.8 MeV).

After eluting the Am fraction from the 12-cm AGMP-1 column (detailed in the curium section), a small amount of the purified Am (3-6 nanograms) was subsequently deposited on a rhenium ribbon filament and analyzed for relative abundances of the americium isotopes by thermal ionization mass spectrometry.

From the alpha analysis (combined with the mass spectrometric analysis), the concentration of the Am isotopes relative to U-238 was calculated.

#### 2.14 Analysis of Np-237

A two milliliter sample of each working solution was spiked with a known quantity of Np-239 tracer ( $t_{1/2} = 2.35$  days). Oxidation states were reduced using an excess of ferrous ammonium sulfate. After 30 minutes, each sample solution was passed through a 2 cm long column of nitrate-form AG1-X4 ion exchange resin. The Np was absorbed by the resin, while fission products, uranium and plutonium were washed off with 7 M nitric acid followed by 0.1 M  $\text{NH}_4\text{I}$  in 12 M HCL. The Np was then eluted from the column and transferred to a stainless steel alpha planchet.

Each alpha planchet was counted by gamma spectrometry to determine the chemical recovery of the added Np-239 "spike". Then the planchets were counted for total alpha count-rate using a calibrated proportional counter. Finally, each planchet was analyzed by alpha spectroscopy to determine the fraction of alpha counts due to Np-237 (4.8 MeV).

Since "excess" Np-239, above the amount added, (produced from the alpha decay of Am-243,  $t_{1/2} = 7370$  years) was found to be present in all samples, a correction to the observed Np-239 yield was made based on the concentration of Am-243 in each sample.

## 2.15 Burnup Determinations

Burnup was calculated for all samples using GE's BRNUP (VAX) code. Burnup was calculated using both our "normal" inputs (U-spiked, Nd-spiked, Pu-spiked and Pu-unspiked) and the addition of the unspiked neodymium mass ratios. (The burnup code is designed to calculate burnup with or without unspiked neodymium values).

Figure 2-1.  
Nitric acid "Gradient" concentration  
(For Rare-earth separations)

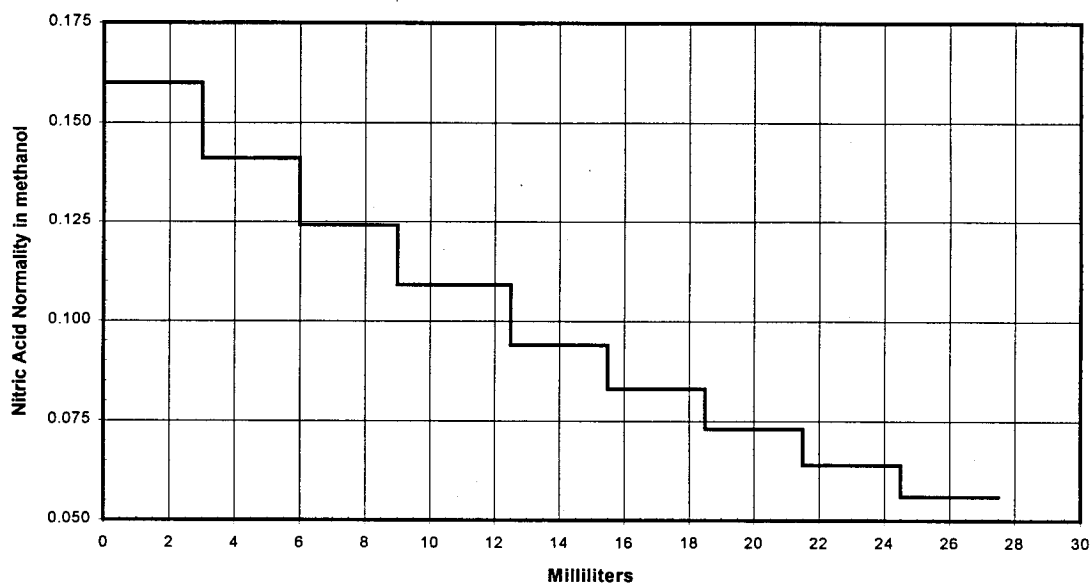


Figure 2-2.  
Rare-Earth Separations on 12-cm AGMP-1 nitrated resin  
Gradient, starting with 1.6 Molar Nitric acid (10% in methanol)

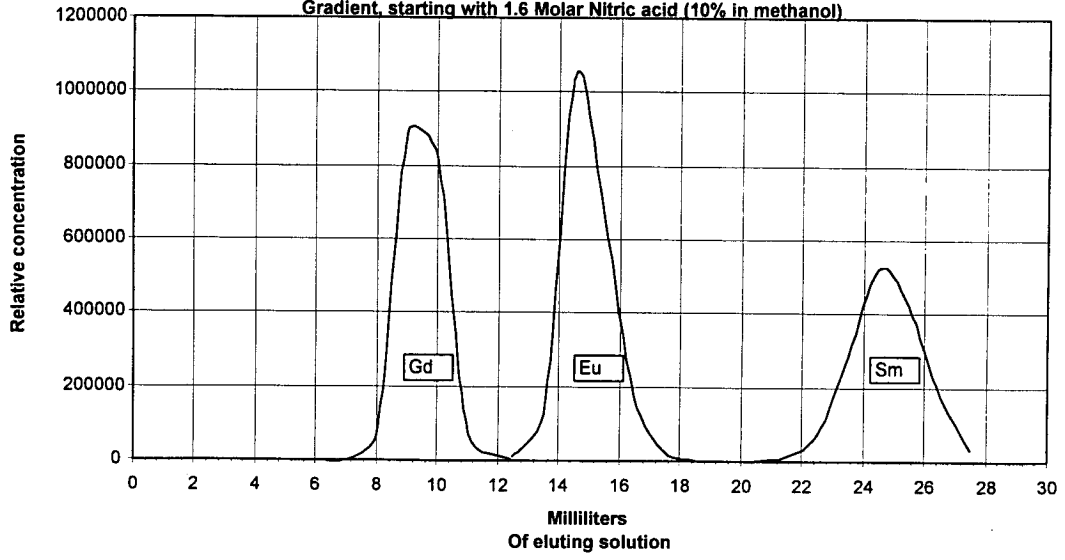


Figure 2-3.  
Nitric acid "Gradient" concentration  
(For Am-Cm separations)

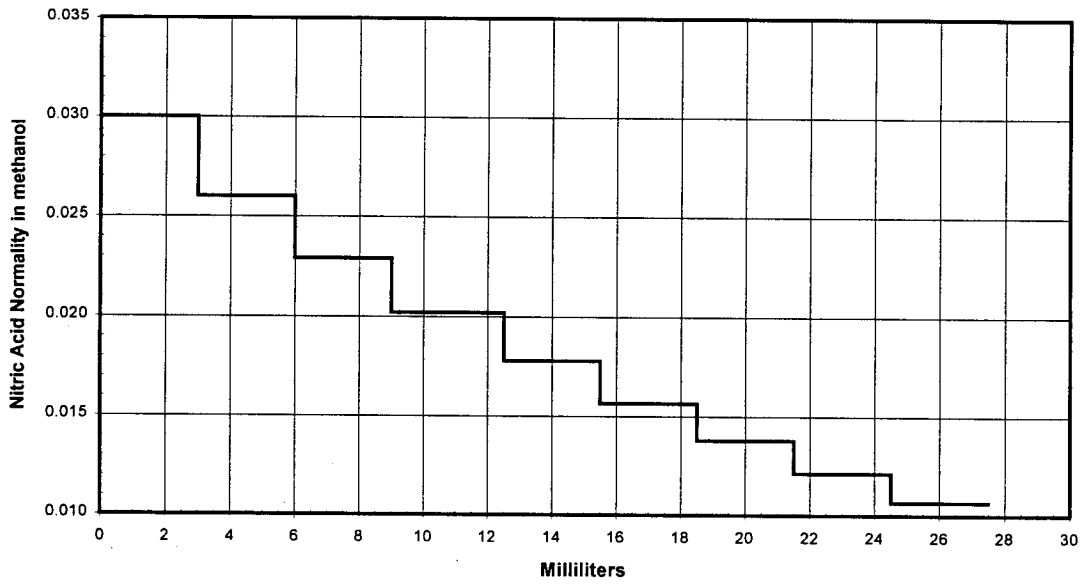
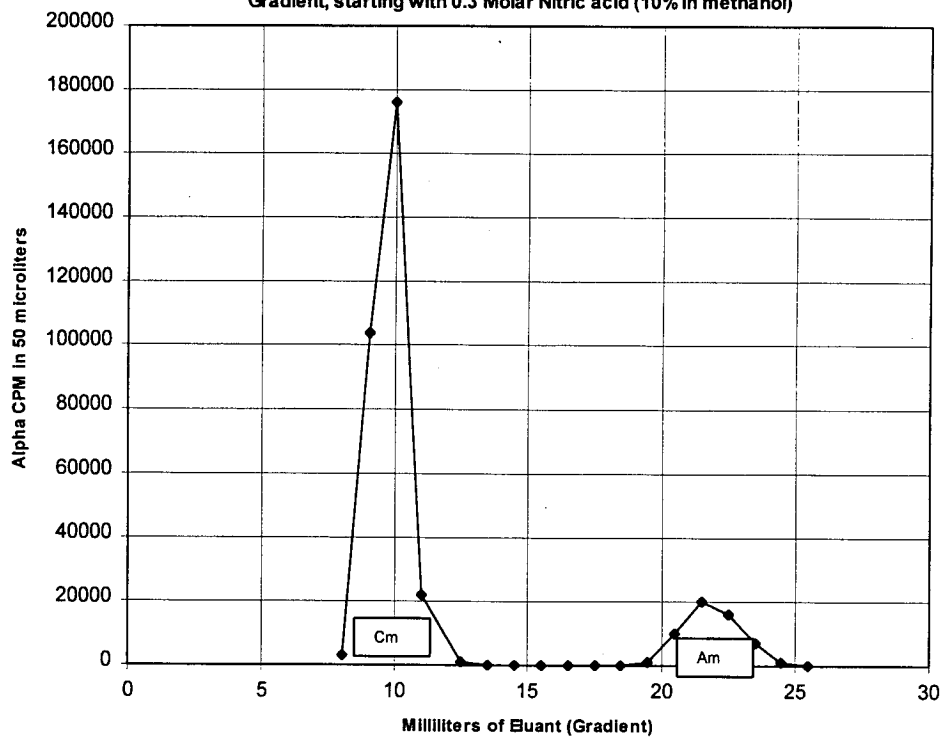


Figure 2-4.

Am and Cm Elution on 12-cm AGMP-1 nitrated resin  
Gradient, starting with 0.3 Molar Nitric acid (10% in methanol)



### 3. Results

The isotopic concentrations and burnup results for the 6 fuel samples investigated under Phase 2 of this contract are shown in Tables 3-1 through 3-11. Table 3-12 summarizes the estimated relative uncertainty, at the 95% confidence level, of each measurement.

**Table 3-1  
Burnup Analysis Summary**

TRW Sample ID	Fuel Rod Identification	Sub-sample	Burnup (MWd/MTU)		
			"Normal" Nd Burnup Anal.	"non-spiked" Nd analysis	Average
2	TMI O1	3A2	2.98E4	2.99E4	2.99E4
5	TMI O12	3A2	2.65E4	2.65E4	2.65E4
7	TMI O13	1A2	2.24E4	2.32E4	2.28E4
8	TMI O13	3A2	2.63E4	2.64E4	2.63E4
9	QC-1 A2	3B1	7.05E4	6.96E4	7.00E4
10	QC-1 A2	5A1B	5.25E4	5.25E4	5.25E4

**Table 3-2  
Uranium Isotope Concentrations**

TRW Sample ID	Fuel Rod Identification	Sub-sample	mg U-234	mg U-235	mg U-236
			Per mg U-238	per mg U-238	per mg U-238
2	TMI O1	3A2	3.25E-04	2.05E-02	5.34E-03
5	TMI O12	3A2	3.34E-04	2.33E-02	4.93E-03
7	TMI O13	1A2	3.65E-04	2.53E-02	4.49E-03
8	TMI O13	3A2	3.40E-04	2.34E-02	4.89E-03
9	QC-1 A2	3B1	1.60E-04	1.25E-03	5.90E-03
10	QC-1 A2	5A1B	1.84E-04	3.86E-03	5.91E-03



**Table 3-3  
Plutonium Isotope Concentrations**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub- sample</u>	<u>mg Pu-238 per mg U-238</u>	<u>mg Pu-239 per mg U-238</u>	<u>mg Pu-240 per mg U-238</u>	<u>mg Pu-241* per mg U-238</u>	<u>mg Pu-242 per mg U-238</u>
2	TMI O1	3A2	1.16E-04	5.98E-03	1.98E-03	9.79E-04	3.04E-04
5	TMI O12	3A2	9.40E-05	6.41E-03	1.76E-03	8.97E-04	2.20E-04
7	TMI O13	1A2	6.41E-05	5.77E-03	1.46E-03	7.04E-04	1.54E-04
8	TMI O13	3A2	9.29E-05	6.28E-03	1.73E-03	8.79E-04	2.16E-04
9	QC-1 A2	3B1	6.66E-04	3.94E-03	3.71E-03	9.98E-04	1.98E-03
10	QC-1 A2	5A1B	5.31E-04	4.47E-03	3.42E-03	1.02E-03	1.23E-03

\*As of 4 Nov. 1999

**Table 3-4  
Neodymium Isotope Concentrations**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub- sample</u>	<u>mg Nd-143 per mg U-238</u>	<u>mg Nd-145 per mg U-238</u>	<u>mg Nd-146 Per mg U-238</u>	<u>mg Nd-148 per mg U-238</u>	<u>mg Nd-150 per mg U-238</u>
2	TMI O1	3A2	8.92E-04	6.87E-04	6.58E-04	3.58E-04	1.64E-04
5	TMI O12	3A2	8.16E-04	6.11E-04	5.76E-04	3.14E-04	1.43E-04
7	TMI O13	1A2	7.41E-04	5.51E-04	5.04E-04	2.77E-04	1.25E-04
8	TMI O13	3A2	8.11E-04	6.08E-04	5.72E-04	3.12E-04	1.42E-04
9	QC-1 A2	3B1	8.77E-04	1.21E-03	1.65E-03	7.79E-04	3.99E-04
10	QC-1 A2	5A1B	9.68E-04	1.03E-03	1.23E-03	6.13E-04	3.06E-04

**Table 3-5  
Cesium Isotope Concentrations**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub-sample</u>	<u>mg Cs-134 per mg of U-238 at count-time*</u>	<u>mg Cs-137 per mg of U-238 at count-time*</u>
2	TMI O1	3A2	2.76E-05	1.17E-03
5	TMI O12	3A2	2.27E-05	1.00E-03
7	TMI O13	1A2	1.76E-05	8.92E-04
8	TMI O13	3A2	2.27E-05	1.01E-03
9	QC-1 A2	3B1	1.85E-05	2.05E-03
10	QC-1 A2	5A1B	1.35E-05	1.62E-03

\* As of 11/5/99

**Table 3-6  
Europium Isotope Concentrations**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub-sample</u>	<u>mg Eu-151 per mg U-238</u>	<u>mg Eu-153 per mg U-238</u>
2	TMI O1	3A2	4.74E-07	1.01E-04
5	TMI O12	3A2	5.02E-07	8.65E-05
7	TMI O13	1A2	4.48E-07	7.13E-05
8	TMI O13	3A2	4.99E-07	8.61E-05
9	QC-1 A2	3B1	5.72E-07	2.34E-04
10	QC-1 A2	5A1B	5.73E-07	1.92E-04

**Table 3-7  
Samarium Isotope Concentrations**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub- sample</u>	<u>mg Sm-147 per mg U-238</u>	<u>mg Sm-149 per mg U-238</u>	<u>mg Sm-150 per mg U-238</u>	<u>mg Sm-151 per mg U-238</u>	<u>mg Sm-152 per mg U-238</u>
2	TMI O1	3A2	2.20E-04	4.36E-06	2.78E-04	1.47E-05	1.07E-04
5	TMI O12	3A2	2.01E-04	4.44E-06	2.41E-04	1.51E-05	9.27E-05
7	TMI O13	1A2	1.86E-04	4.23E-06	2.06E-04	1.35E-05	8.47E-05
8	TMI O13	3A2	1.99E-04	4.42E-06	2.38E-04	1.51E-05	9.19E-05
9	QC-1 A2	3B1	3.96E-04	1.40E-06	5.23E-04	1.01E-05	1.92E-04
10	QC-1 A2	5A1B	3.86E-04	1.76E-06	4.23E-04	1.02E-05	1.59E-04

**Table 3-8  
Gd-155 Concentrations**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub- sample</u>	<u>mg Gd-155 per mg U-238</u>
2	TMI O1	3A2	3.09E-06
5	TMI O12	3A2	2.68E-06
7	TMI O13	1A2	2.10E-06
8	TMI O13	3A2	2.70E-06
9	QC-1 A2	3B1	1.28E-05
10	QC-1 A2	5A1B	9.97E-06

**Table 3-9  
Curium Isotope Concentrations**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub- Sample</u>	<u>(Alpha Spec) mg Cm-242* per mg U-238</u>	<u>(Mass Spec) mg Cm-242* per mg U-238</u>	<u>mg Cm-243 per mg U-238</u>	<u>mg Cm-244 per mg U-238</u>	<u>mg Cm-245 per mg U-238</u>
2	TMI O1	3A2	1.2E-08	2.3E-08	1.25E-07	7.68E-06	4.02E-07
5	TMI O12	3A2	1.1E-08	1.3E-08	1.07E-07	5.51E-06	2.90E-07
7	TMI O13	1A2	7.4E-09	7.5E-09	5.97E-08	2.62E-06	1.14E-07
8	TMI O13	3A2	1.1E-08	1.4E-08	1.01E-07	5.23E-06	2.74E-07
9	QC-1 A2	3B1	ND	ND	1.58E-06	2.47E-04	1.46E-05
10	QC-1 A2	5A1B	ND	ND	1.31E-06	1.01E-04	6.70E-06

\* As of 26 Nov. 1999  
ND =Not Detected

**Table 3-10  
Americium Isotope Concentrations**

<u>TRW Sample ID</u>	<u>Fuel Rod Identification</u>	<u>Sub- sample</u>	<u>mg Am-241 per mg U-238</u>	<u>mg Am-242m per mg U-238</u>	<u>mg Am-243 per mg U-238</u>
2	TMI O1	3A2	2.12E-04	4.53E-07	3.75E-05
5	TMI O12	3A2	2.22E-04	5.18E-07	2.96E-05
7	TMI O13	1A2	1.73E-04	3.36E-07	1.71E-05
8	TMI O13	3A2	2.16E-04	4.99E-07	2.85E-05
9	QC-1 A2	3B1	4.72E-04	1.35E-06	4.87E-04
10	QC-1 A2	5A1B	4.86E-04	1.90E-06	2.58E-04

**Table 3-11**  
**Np-237 Concentrations**

<u>TRW</u> <u>Sample ID</u>	<u>Fuel Rod</u> <u>Identification</u>	<u>Sub-</u> <u>sample</u>	<u>mg Np-237</u> <u>per mg U-238</u>
2	TMI O1	3A2	4.23E-04
5	TMI O12	3A2	3.72E-04
7	TMI O13	1A2	3.01E-04
8	TMI O13	3A2	3.71E-04
9	QC-1 A2	3B1	7.82E-04
10	QC-1 A2	5A1B	6.78E-04

**Table 3-12**  
**Estimated Measurement Uncertainties**

Isotope*	Estimated percentage uncertainty @95% confidence
U-238	1
U-234	1
U-235	1
U-236	1
Cs-134	3.5
Cs-137	3.5
Pu-238	5
Pu-239	1.2
Pu-240	1.2
Pu-241	1.2
Pu-242	1.2
Nd-143	1.5
Nd-145	1.5
Nd-146	1.5
Nd-148	1.5
Nd-150	1.5
Sm-147	1.7
Sm-149	1.8
Sm-150	1.7
Sm-151	1.7
Sm-152	1.7
Eu-151	1.7
Eu-153	1.8
Gd-155	2.7**
Am-241	7
Am-242	7
Am-243	7
Cm-242	20
Cm-243	5.5
Cm-244	5.5
Cm-245	5.5
Np-237	5.8

\*reported as mg/mg U-238 (except for the U-238 measurement)

\*\*Interference

#### 4.0 Reference Materials

1. Burnup "Triple-spike" - - "0-3-2"  
Contains: 2.726x10<sup>16</sup> Atoms U-233/ml  
3.489x10<sup>16</sup> Atoms Pu-242/ml  
1.817x10<sup>15</sup> Atoms Nd-150/ml
2. Gd-158 - - ORNL Batch 109690  
97.5% Isotopically Pure
3. Sm-152 - - ORNL Batch 167692  
99.4% Isotopically Pure
4. Eu-151 - - ORNL Batch 151892  
96.8% Isotopically Pure
5. Gadolinium, Standard, 10,000 µg/ml  
Traceable to NIST SRM 928  
Lot No. ID-0887  
ULTRA Scientific
6. Samarium Standard, 10,000 µg/ml  
Traceable to NIST SRM 3147  
Lot No. ID-0641  
ULTRA Scientific
7. Europium Standard, 10,000 µg/ml  
Traceable to NIST SRM 3117  
Lot No. ID-0637  
ULTRA Scientific
8. Plutonium Isotopic Standard, SRM 946  
NIST
9. Uranium Isotopic Standards, U010 and U050  
NIST

**5. Equipment List**

1. Thermal Ionization Mass Spectrometer

Finnigan MAT  
Model 261

2. Gamma Spectrometer

Nuclear Data 6700  
Ortec HPGe detector

3. Proportional Counter

NMC  
Model PC-5

4. Alpha Spectrometer

Ortec  
Model 676A  
Plus GENIE 2000 Analyzer (Canberra)