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Benchmark Data for Validating Irradiated Fuel Compositions Used in Criticality Calculations

S. R. Bierman
R. J. Talbert

October 1994

Prepared for the U.S. Department of Energy
Office of Civilian Radioactive
Waste Management
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
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Summary

To establish criticality safety margins utilizing burnup credit in the storage and transport of spent reactor fuels requires a knowledge of the uncertainty in the calculated fuel composition used in making the reactivity assessment. To provide data for validating such calculated burnup fuel compositions, radiochemical assays have been obtained as part of the United States Department of Energy From-Reactor Cask Development Program. Assay results and associated operating histories on the initial three samples analyzed in this effort are presented. The three samples were taken from different axial regions of a Pressurized Water Reactor fuel rod and represent radiation exposures of about 37, 27, and 44 GWd/MTU. The data are presented in a benchmark type format to facilitate identification/referencing and computer code input.

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1.0 Introduction

Traditionally, criticality analyses of nuclear fuel storage and transport have assumed the systems contained unirradiated fuel. This assumption has been accepted as providing a significant criticality safety design margin without incurring undue costs and restrictions since operations were generally limited by heat transfer and shielding considerations. However, as irradiated fuels are aged out-of-reactors, criticality considerations begin to limit design and operations. The results of a study (Sanders, 1987) have indicated that considerable safety benefits and cost savings can be achieved in the transport of aged spent fuel if credit is taken for burnup (irradiation/cooling history) in the criticality design analyses of fuel transport systems.

To establish the criticality safety margin associated with a transport design utilizing burnup credit requires a knowledge of the uncertainty in the calculated fuel composition used in making the reactivity assessment. To provide data for validating such calculated burnup compositions, radiochemical assays have been obtained on a representative reactor fuel having experienced irradiation exposures up to about 44 GWd/MTU. The data are presented in benchmark type format to facilitate identification/referencing and computer code input. The narrative associated with each benchmark is kept to the minimum required to clarify the data and identify its source.

Each sample assay consists of a set of radiochemical analytical data obtained by destructively assaying a spent fuel rod that had experienced a unique set of operating conditions. The unique operating conditions associated with each sample analysis are specified to the extent required by current burnup calculational techniques. Also, the unirradiated composition and the source of the fuel rod sampled are presented. All of the samples destructively assayed were located away from any fuel assembly hardware such as end fittings and grid spacer plates. The presence or location of other perturbations such as water holes, fuel assembly boundaries, and burnable poisons are identified as part of the operating conditions.

2.0 Radiochemical Procedures

The analytical procedures used for sample preparation, determining burnup, and analyzing for the indicated isotopic concentrations are briefly covered below. An estimate of the one standard deviation error limits on the analytical results obtained by each procedure is also provided.

Analysis	Description
Sample Preparation	Weighed sample is dissolved in heated 12N HNO ₃ (+ trace HF). Solution is separated from cladding and made up to 100 mL. Aliquots are taken for subsequent analysis. Uncertainty associated with sample preparation: ±1.0%.
Burnup (including U and Pu isotopes)	Fission product neodymium is chemically separated from irradiated fuel and determined by isotopic dilution mass spectrometry. Enriched ¹⁵⁰ Nd is used as the ND isotope diluent, and mass 142 is used to determine natural Nd contamination. Uranium and plutonium are also determined by mass spectrometry. The method uses a calibrated triple spike of ¹⁵⁰ Nd, ²³³ U, and ²⁴² Pu per ANSI/ASTM Standard Test Method E-321-79. Total uncertainty: Atom % burnup, ±2.5%; Pu, ±1.6%; U, ±1.6%.
²⁴¹ Am	Americium is separated, along with curium, using cation and anion exchange and determined by alpha spectrometry. Uncertainty: ²⁴¹ Am, ±4.9%.
¹³⁵ Cs	Cesium is separated from other elements by chromatographic elution from a cation exchange column. Isotopic abundance of the cesium isotope is determined by mass spectrometry. Uncertainty: ±14%.
¹³⁷ Cs	The cesium is determined by gamma ray spectrometry on an aliquot of the aqueous solution. Uncertainty: ±3.5%.
⁹⁹ Tc	Technetium is separated from other radioactive species by a process that absorbs most of the other species onto a cation exchange resin. The technetium is extracted from the effluent into hexone as tetraphenylarsonium pertechnetate. The technetium activity is then measured by beta counting. Uncertainty: ±3.5%.
²³⁷ Np	Neptunium-237 is separated from other radionuclides species by extraction into a mixture of tri-iso-octylamine (TiOA) in xylene, stripped from the TiOA phase with HCl and re-extraction into a mixture of thenolyl-trifluoro acetone (TTA) in xylene for additional separation. Neptunium-237 is measured by alpha counting. A ²³⁹ Np tracer is added to the sample and gamma-counted to determine a recovery factor. Uncertainty: ±1.9%.
Lanthanides (except Nd)	The lanthanide elements are separated from the dissolved fuel sample by extraction chromatography followed by cation exchange per Pacific Northwest Laboratories Technical Procedure PNL-ALO-498. (Procedure developed for these assays. See Appendix A for more detailed information.) Individual lanthanide concentrations are determined from Inductively Coupled Plasma-Mass Spectrometry measurements relative to ¹⁴⁵ Nd and ¹⁴³ Nd. Uncertainty: See Appendix B.

3.0 Benchmark Data

Radiochemical assays of Pressurized Water Reactor fuel samples having experienced irradiations of 27.35, 37.12, and 44.34 GWd/MTU are presented in this benchmark. The three samples were obtained from different axial locations in a single fuel rod corresponding to maximum burnup and approximately 85% and 60% of maximum burnup. Thus, the benchmark provides composition data as a function of fuel burnup and data on the axial dependency of burnup within the same rod.

3.1 Unirradiated and Irradiated Fuel Assays

As indicated in the unirradiation description of the fuel samples, the samples were obtained from a Combustion Engineering fuel assembly. The assembly had experienced four irradiation cycles in the Calvert Cliff Unit No. 1 reactor. Except for the lanthanides, the samples had been previously analyzed by the Department of Energy's Materials Characterization Center (MCC) at the Pacific Northwest Laboratories. Results of these MCC analyses that are of interest to criticality calculations are included as part of the chemical compositions presented herein. The unirradiated fuel data and fuel assembly information was provided by Combustion Engineering and by Baltimore Gas and Electric Company (Combustion Engineering, 1986; Baltimore Gas and Electric Company, 1968). Additional information can be obtained from PNL-5109-104 (Guenther, 1991).

As stated in Section 2, the lanthanide isotope concentrations, except for the neodymiums, were determined by mass spectrometry measurements. As a consequence, irradiated concentrations for four of the nuclides of interest to criticality calculations are based on measurement data contaminated with significant quantities of nuclides of more than one element. These are nuclides having mass numbers 147 (Pm, Sm), 150 (Nd, Sm), 151 (Sm, Eu) and 155 (Eu, Gd). These measurement data have been adjusted using calculated values to obtain the concentrations shown in the table on irradiated compositions. The measurement data and the associated adjustments made to these four sets of data are presented in the paragraphs that follow. The mass spectrometry measurement data for all the lanthanides can be found in Appendix C.

Based on calculations,^(a) about 3.48% of the isobar for mass number 147 was due to

(a) ORIGEN-S calculated results using SAS2 27-group ENDF/B4 library. Facsimile communication of only the results received from C. V. Parks, Oak Ridge National Laboratory, June 18, 1993.

Pm in the 27.35 GWd/MTU spent fuel sample. Similarly, the 37.12 GWd/MTU and the 44.34 GWd/MTU samples contained about 3.49 wt% Pm and 3.51 wt% Pm, respectively. The measured concentration of mass number 147 in the three samples (0.221 mg/gUO₂, 0.254 mg/gUO₂, and 0.268 mg/gUO₂, respectively) were corrected, using a half life of 2.623 yr, for change in Pm content from June, 1987 to the time of measurement (October, 1992) to obtain the ¹⁴⁷Sm concentrations given in the table on irradiated compositions.

As stated above, ¹⁵⁰Nd concentrations in the samples were obtained by a second independent measurement method. The values given in the table for ¹⁵⁰Sm were obtained by taking the difference between these independently measured data and the mass spectrometry measurement data for mass number 150. Although the ¹⁵⁰Nd is not of interest to criticality calculations, the independently measure value is given in the table for completeness.

Both components (Sm and Eu) in the isobar for mass number 151 are essentially stable relative to the time difference between June 1987 and the time of measurement (October, 1992). Consequently, the measured isobaric data were adjusted based on calculated values^(a) of 7.95 wt%, 7.89 wt%, and 7.85 wt% Eu in the isobaric data, respectively, for the 27.35 GWd/MTU, the 37.12 GWd/MTU and the 44.34 GWd/MTU samples. The total concentration for mass number 151 in each of the three samples was measured at 0.934E-02 mg/gUO₂, 0.930E-02mg/g UO₂ and 0.978E-02 mg/gUO₂, respectively.

Calculations^(a) indicate the isobar for mass number 155 contained about 20.99 wt% Eu for the 27.35 GWd/MTU sample, about 21.05 wt% for the 37.12 GWd/MTU sample and about 21.09 wt% for the 44.34 GWd/MTU sample. As with mass number 147, the measured concentrations (0.474E-02 mg/gUO₂, 0.710E-02 mg/gUO₂ and 0.982E-02 mg/gUO₂, respectively) for mass number 155 were adjusted using these calculated values and corrected from June, 1987 to the time of measurement (October, 1992) using a half life of 4.68 yr for ¹⁵⁵Eu, to obtain the ¹⁵⁵Gd and the ¹⁵⁵Eu concentrations given in the table on irradiated compositions.

Except for the lanthanide elements, estimated one sigma error limits are given in Section 2 for the measurement results presented in Table 3.2. To obtain data for estimating the error limits associated with the lanthanides, sample 87-81 was analyzed by the Los Alamos National Laboratory (LANL) and the Khlopin Radium Institute (KRI) in

(a) ORIGEN-S calculated results using SAS2 27-group ENDF/B4 library. Facsimile communication of only the results received from C. V. Parks, Oak Ridge National Laboratory, June 18, 1993.

addition to the Pacific Northwest Laboratories (PNL). One sigma standard deviation error limits (in units of weight %) for each of the lanthanides are summarized below:

M-155-29% ^(b)

Eu-153-2%

Sm-152-3%

Sm-151-7%

Sm-150-2%

Sm-149-18%

Sm-147-4%

Nd-145<1%

Nd-143<1%

Analyses of the three sets of data to obtain the above estimated error limits are presented in Appendix B. The complete LANL measurement data are presented in Appendix D. The complete KRI measurement data are presented in Appendix E. The KRI measurements include mass spectrometry data before and after chemical separation of the lanthanides into individual elements. The LANL and PNL measurements involved only mass spectrometry measurements without chemical separation into elemental lanthanides.

Some of the earlier spent fuel analyses data were reported in curies per gram of fuel by the Material Characterization Center in PNL-5109-104 (Guenther, 1991). For inclusion in Table 3.1, these analytical results have been converted to mg/g fuel using the respective half-life values as indicated below:

ISOTOPE	HALF LIFE
99-Tc	2.1E + 05y
135-Cs	2.3E + 06y
137-Cs	30.2y
237-Np	2.14E + 06y
241-Am	432y

^(b)Mass isobar at mass 155

Table 3.1 Source and Unirradiated Composition of Samples

Reactor	Calvert Cliffs No. 1
Fuel Assembly	Combustion Engineering D047
Assembly Spacing, cm	20.78, center-to-center
Assembly Cross Section, cm	20.27 x 20.27, outside fuel rods
Fuel Rod Arrangement	14 x 14, 1.473 cm center to center
Fuel Rod	MKP109
Approved Testing Material No.	ATM104 ^(a)
Fuel Pellet	
Material	UO ₂ (CE Lot B-71-GB)
Diameter, cm	0.9563
Length, cm	1.143
Density, g/cm ³	10.36-10.48
Grain size, μ m	≥ 5
Unirradiated Fuel Composition	
Uranium, wt%	88.143 $\pm$ 0.010
O:U Atom Ratio	2.001 $\pm$ 0.002
²³⁴ U, wt%	0.025 ^(b)
²³⁵ U, wt%	3.038
²³⁶ U, wt%	0.025 ^(b)
Carbon, ppm	17.5 $\pm$ 6.1
Nitrogen, ppm	23.2 $\pm$ 8.3
Fluorine, ppm	<5
Chlorine and Fluorine, ppm	<10
Iron, ppm	<45
Silver, ppm	<1
Aluminum, ppm	<115
Silicon	--
Nickel	<25
Cladding	
Material	Zircaloy-4 (Sandvik Lot 5FP65)
Outside Diameter, cm	1.118
Inside Diameter, cm	0.986
Density, g/cm ³	6.32 ^(b)

(a) See PNL-5109-104, Guenther, 1991.

(b) Nominal values, not part of fuel certification data.

Table 3.2 Irradiated Composition^(a) of Fuel and Total Burnup - Fuel Assembly D047, Rod MKP109

Sample	87-81	87-72	87-63
Axial Location, ^(b) cm	13.20	27.70	165.22
Burnup, ^(c) GWd/MTU	27.35	37.12	44.34
²³⁴ U, mg/g fuel	0.16	0.14	0.12
²³⁵ U, mg/g fuel	8.47	5.17	3.54
²³⁶ U, mg/g fuel	3.14	3.53	3.69
²³⁸ U, mg/g fuel	842.50	832.70	824.86
²³⁸ Pu, mg/g fuel	0.101	0.189	0.269
²³⁹ Pu, mg/g fuel	4.264	4.357	4.357
²⁴⁰ Pu, mg/g fuel	1.719	2.239	2.543
²⁴¹ Pu, mg/g fuel	0.681	0.903	1.020
²⁴² Pu, mg/g fuel	0.289	0.576	0.840
²³⁷ Np, mg/g fuel	0.268	0.356	0.468
²⁴¹ Am, mg/g fuel	0.249	0.343	0.381
¹³³ Cs, mg/g fuel	0.85	1.09	1.24
¹³⁵ Cs, mg/g fuel	0.36	0.40	0.43
¹³⁷ Cs, mg/g fuel	0.77	1.04	1.25
¹⁴³ Nd, mg/g fuel	0.613	0.716	0.763
¹⁴⁴ Nd, mg/g fuel	0.943	1.338	1.643
¹⁴⁵ Nd, mg/g fuel	0.510	0.653	0.744
¹⁴⁶ Nd, mg/g fuel	0.490	0.682	0.830
¹⁴⁸ Nd, mg/g fuel	0.265	0.359	0.428
¹⁵⁰ Nd, mg/g fuel	0.124	0.172	0.208
¹⁴⁷ Sm, mg/g fuel	0.190	0.218	0.230
¹⁴⁸ Sm, mg/g fuel	0.106	0.164	0.222
¹⁴⁹ Sm, mg/g fuel	0.0029	0.0030	0.0047
¹⁵⁰ Sm, mg/g fuel	0.207	0.271	0.361
¹⁵¹ Sm, mg/g fuel	0.0086	0.0086	0.0090
¹⁵² Sm, mg/g fuel	0.087	0.104	0.121
¹⁵¹ Eu, mg/g fuel	0.0007	0.0007	0.0008
¹⁵³ Eu, mg/g fuel	0.079	0.109	0.148
¹⁵⁵ Eu, mg/g fuel	0.0021	0.0033	0.0045
¹⁵⁵ Gd, mg/g fuel	0.0025	0.0039	0.0053
⁹⁹ Tc, mg/g fuel	0.56	0.72	0.78

(a) Radiochemical analyses performed during June 1987, or corrected for decay to June 1987.

(b) From bottom of fuel.

(c) Burnup based on measured ¹⁴⁸Nd concentration (ASTM 1985).

3.2 Power History and Boron Concentration-Fuel Assembly D047, Rod MKP109

The power histories and axial distributions reported for each reactor cycle in the table that follows were obtained from reactor operating records. For each operating cycle, time dependent axial power distribution data were available on each fuel assembly along with the assembly average power density. However, only time dependent average power densities existed for individual fuel rods in each fuel assembly. The time dependent average power densities for the specific fuel rod (MKP109) and the time dependent average power density for fuel assembly (D047) were used to ratio that fuel assembly's axial power density distribution to obtain a local heat generation rate for each of the axial locations at which sample analyses data are presented.

As indicated in the power history tables, each irradiation cycle is divided into time intervals of differing lengths. The operating records report these simply as time points. To calculate the length of each interval in days, the initial metal density of the fuel, the rod burnup over a given time interval and the average linear heat generation rate in the rod for that time interval were used.

The average soluble boron concentration for each time interval during a reactor cycle was also obtained from reactor operating records. The value given for each time interval in the tables that follow is the arithmetic mean of the boron concentration at the beginning and end of the time interval.

A ^{137}Cs gamma scan of rod MKP109 is also presented in this section to illustrate the axial irradiation experienced by the fuel rod and to indicate the axial locations of the samples referred to in Section 3.1. The sampling points are identified as P, CC and LL. The gamma scan also provides a means of predicting axial burnup in the fuel rod. It has been demonstrated (Guenther, 1991) that the ^{137}Cs activity is a linear function of fuel burnup. The linear relationship determined (Guenther, 1991) for rod MKP109 is:

$$\text{BU} = 0.0222 \text{ A} - 6.8,$$

where A is the measured activity (counts per second) presented in the gamma scan and BU (MWd/MTU) is the burnup at any axial location of rod MKP109. Thus the gamma scan can be used to develop an axial burnup profile if desired.

**Table 3.3 Power Histories and Boron Concentrations-Fuel Assembly D047,
Rod MKP109**

CYCLE No. 2 3-22-77 to 1-22-78					CYCLE No. 3 4-3-78 to 4-20-79				
Interval days	Average Soluble Boron, ppm	LHGR ^(a) kW/ft			Interval days	Average Soluble Boron, ppm	LHGR ^(a) kW/ft		
		13.20 cm	27.70 cm	165.22 cm			13.20 cm	27.70 cm	165.22 cm
7.0	654	2.0	3.04	5.42	7.9	883	2.63	3.85	7.47
30.8	614	2.73	4.08	6.64	14.4	862	2.71	3.95	7.20
16.3	563	2.80	4.18	6.60	19.7	837	2.14	3.11	5.04
11.4	533	2.86	4.26	6.60	16.8	808	3.18	4.58	7.79
12.5	507	2.94	4.34	6.56	16.3	775	3.35	4.81	7.78
23.7	468	3.01	4.43	6.41	15.4	741	3.74	5.33	8.16
22.8	418	3.16	4.61	6.44	39.1	684	3.71	5.23	7.45
23.2	368	3.27	4.73	6.37	31.2	611	3.95	5.50	7.37
8.1	333	3.30	4.75	6.34	31.8	545	4.08	5.64	7.20
31.4	290	3.48	4.95	6.34	31.8	478	4.20	5.75	7.02
34.3	218	3.59	5.05	6.53	44.3	368	4.38	5.93	6.93
16.4	162	3.75	5.25	6.38	25.0	291	0	0	0
19.2	122	3.75	5.21	6.26	59.1	224	4.67	6.20	6.89
12.8	86	3.81	5.28	6.30	28.9	120	4.95	6.52	7.12
34.2	36	3.63	4.98	5.95	81	83	0	0	0
1.9	0	3.64	4.98	5.96					
71	0	0	0	0					

CYCLE No. 4 7-10-79 to 10-18-80					CYCLE No. 5 1-11-81 to 4-17-82				
Interval days	Average Soluble Boron, ppm	LHGR ^(a) kW/ft			Interval days	Average Soluble Boron, ppm	LHGR ^(a) kW/ft		
		13.20 cm	27.70 cm	165.22 cm			13.20 cm	27.70 cm	165.22 cm
46.1	960	2.79	3.97	6.59	65.0	919	1.98	2.67	4.55
24.0	889	2.91	4.11	6.54	5.5	911	2.15	2.91	4.80
22.6	827	3.07	4.29	6.43	6.6	896	2.20	2.98	4.80
25.7	759	3.29	4.56	6.33	28.6	854	2.36	3.15	4.79
30.2	706	1.57	2.17	3.05	31.2	784	2.54	3.36	4.72
41.2	788	1.79	2.46	3.18	27.0	715	2.66	3.52	4.73
50.3	720	1.72	2.35	3.00	22.7	655	2.77	3.63	4.71
11.0	673	3.18	4.33	5.23	27.1	603	2.46	3.22	4.05
32.8	527	3.93	5.31	6.17	55.2	521	2.81	3.64	4.34
23.5	460	4.07	5.42	6.12	20.9	434	3.17	4.08	4.75
29.4	370	4.05	5.35	5.95	41.9	356	3.32	4.24	4.79
28.1	301	4.25	5.56	6.07	21.6	281	3.38	4.30	4.80
65.4	191	4.42	5.67	5.98	27.6	226	3.21	4.04	4.49
35.7	73	4.62	5.87	6.02	19.0	173	3.57	4.49	4.87
85	31	0	0	0	61.2	79	3.05	3.79	4.00
					1870 ^(b)	-	0	0	0

(a) At indicated axial locations from bottom of fuel.

(b) Cooling time to date of sample analyses (June 1987).

Figure 3.1 Gamma Scan of Rod MKP109 with Sampling Locations Indicated

3.3 Operating Data - Fuel Assembly D047, Rod MKP109

Nominal operating conditions during each of the four cycles that the assembly was in the reactor are given in the table that follows. Calculated (Cunningham, 1984) temperatures of the fuel at each axial location sampled are also given in the table for each irradiation cycle.

For general information, the locations of fuel assembly D047 in the reactor during the four irradiation cycles are identified in this section also. Also identified in this section is the location of fuel rod MKP109 in the assembly.

Table 3.4 Operating Data - Fuel Assembly D047, Rod MKP109

Cycle Number	<u>2</u>	<u>3</u>	<u>4</u>	<u>5</u>
Cycle Burnup, GWd/MTU	8.117	10.491	10.984	10.811
Nominal Coolant Temperature, °C ^(a)				
Reactor Inlet	288	288	288	288
Reactor Outlet	344	344	344	344
Nominal Coolant Pressure, psia ^(a)				
Reactor Inlet	2250	2250	2250	2250
Reactor Outlet	N.A.	N.A.	N.A.	N.A.
Effective Fuel Temperature, °C ^(b)				
13.02 cm	556	577	502	436
27.70 cm	667	654	520	439
165.22 cm	724	685	521	474

-
- (a) Design parameters per USAEC Docket No. 50-317 (Baltimore Gas and Electric Co., 1986). Personal communication on operating conditions give values of 284C, 312C, and 2247 psia.
- (b) Time weighted volume averaged fuel temperatures at indicated axial locations from bottom of fuel as calculated using computer code GT2R2 (Cunningham, 1984).

Figure 3.2 Location of Fuel Assembly in Reactor Core - Calvert Cliffs No. 1

*Bracketed Numbers Indicate the Cycle of Operation at the Indicated Location

Figure 3.3 Location of Fuel Rod in Fuel Assembly -
Combustion Engineering Fuel Assembly D047

A	MKP 033	MKP 030	MKP 061	MKP 007	MKP 034	MKP 043	MKP 081	MKP 039	MKP 067	MKP 080	MKP 079	MKP 092	MKT 001*	MKP 029
B	MKP 047	MKP 051	MKP 032	MKP 017	MKP 073	MKP 045*	MKP 063	MKP 106	MKP 049	MKP 118	MKP 116	MKT 033	MKP 124	MKT 023*
C	MKN 011	MKN 160	Guide Tube		MKP 119	MKN 047	MKP 103	MKP 054	MKP 056	MKP 112	Guide Tube		MKU 061*	MKP 025
D	MKN 002	MKP 001			MKP 042	MKN 033	MKP 070	MKP 087	MKP 045	MKP 108			MKP 090	MKU 014*
E	MKN 150	MKP 019	MKP 014	MKP 104	MKP 110	MKP 150	MKP 083	MKP 103	MKP 026	MKP 077	MKT 011*	MKP 115	MKU 065*	MKP 048
F	MKN 139	MKP 006*	MKP 018	MKP 005	MKP 057	MKP 060	MKP 059	MKP 068	MKP 101	MKP 096	MKP 097	MKP 002*	MKP 089*	MKT 006*
G	MKP 003	MKN 073	MKP 013	MKP 072	MKP 050	MKP 020	Guide Tube		MKP 069	MKP 100	MKT 015*	MKP 120	MKT 058*	MKP 028
H	MKN 153	MKP 021	MKP 046	MKP 075	MKP 015	MKP 011			MKP 093	MKT 017*	MKP 125	MKT 007	MKP 044	MKT 019*
I	MKP 002	MKN 144	MKP 035	MKP 027	MKP 008	MKP 041	MKP 038	MKP 058	MKP 104	MKP 111	MKT 020	MKP 121	MKU 062	MKP 053
J	MKP 076	MKP 071	MKP 009	MKP 036	MKP 004	MKP 010	MKP 086	MKP 126	MKP 085	MKT 061*	MKP 102	MKT 022*	MKP 127	MKT 021*
K	MKP 037	MKP 040	Guide Tube		MKT 004*	MKP 062	MKT 003*	MKP 117	MKT 013	MKP 105	Guide Tube		MKU 051*	MKP 024
L	MKP 184	MKT 018*			MKP 022	MKT 027*	MKP 052	MKT 005*	MKP 084	MKT 010*			MKP 082	MKT 030*
M	MKU 025*	MKP 016	MKT 014*	MKP 031	MKU 010*	MKP 066*	MKU 011*	MKP 065	MKU 063	MKP 074	MKU 013*	MKP 122	MKU 064*	MKP 012
N	MKN 169	MKT 008*	MKP 023	MKU 050*	MKP 078	MKT 028*	MKP 064	MKT 009*	MKP 107	MKT 035*	MKP 091	MKT 029*	MKP 099	MKT 012*
	1	2	3	4	5	6	7	8	9	10	11	12	13	14

Note: Fuel rod serial numbers are presented as manufactured. Those indicated by * were replaced with special fuel rods for one or more cycles to study fuel and cladding structural properties (Ruzauskas, 1985). Rod MKP 045 was replaced after the fourth cycle of reactor operations with a 2.82 w/o ²³⁵U enriched UO₂ rod (AHS 077, EPRI Test Rod 38) containing a pore former to study in-reactor densification.

4.0 Conclusions and Recommendations

The chemical composition of three spent PWR fuel samples have been analyzed to provide measured chemical compositions of interest in nuclear criticality calculations. The samples were taken at three different axial locations in a single fuel rod corresponding to measured burnups of 27.35 GWd/MTU, 37.12 GWd/MTU and 44.34 GWd/MTU. One sample (27.35 GWd/MTU) was analyzed by three different laboratories to establish estimated error limits for the strong neutron absorbing non-actinide nuclides. It is recommended that the other two samples be analyzed by at least a second laboratory to better establish error limits over the entire range of burnup currently of primary interest to storage and shipment of spent reactor fuel. It is also recommended that samples from a different spent fuel rod be similarly analyzed to provide improved confidence in the measurement data and future criticality calculations. These samples should also be analyzed for rhodium, which is one of the strong neutron absorbers in spent reactor fuel. The initial three samples analyzed were not analyzed for rhodium, in that no established method for rhodium analysis has been developed. Future work will incorporate an initiative to separate and analyze for rhodium.

5.0 References

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APPENDIX A

Separation and Analyses of Lanthanide Component in Spent Fuel

APPENDIX A

Separation and Analyses of Lanthanide Component in Spent Fuel

Except for the lanthanides, the chemical assay data presented in Table 3.2 of this report were obtained using established and well-documented procedures as identified in Section 2.0, Radiochemical Procedures. The procedure used in obtaining the data presented in Table 3.2 for the lanthanide elements consisted of a combination of existing measurement techniques. Although each of the measurement methods used in the lanthanide analyses are currently accepted analytical techniques, their adaptation to analyzing spent reactor fuel are briefly described in this appendix.

Since spent reactor fuel consists of a multitude of chemical elements, the approach taken in analyzing for lanthanide isotopes was to first dissolve a fuel sample in nitric acid, chemically separate the lanthanide component from the spent fuel sample, and analyze this component for the lanthanide isotopes of interest to nuclear criticality by mass spectrometry measurements. The isotopes of interest are strong neutron absorbers and consist of nuclides having mass numbers of 147 (samarium), 150 (samarium and neodymium), 151 (samarium and europium), and 155 (europium and gadolinium). To permit quantifying the results, the mass spectrometry of the neodymium isotopes were determined by the ANSI/ASTM Standard Test Method as indicated in Section 2 of the report.

Following dissolution of the fuel in nitric acid, extraction chromatography and ion exchange are used to isolate the lanthanide fission products from the spent fuel samples. Aliquots, containing up to 10 mg of uranium, are taken from each fuel sample for lanthanide element separation. Uranium and tetravalent actinides are separated from each solution sample by passing a sample through a U/TEVA-Spec* resin column (* registered trademark of EIChroM Industries). The uranium and tetravalent actinides are retained on the resins. The lanthanides and americium pass through the column into a RE-Spec* resin column where the lanthanides (rare earths) and the americium are retained (* registered trademark of EIChroM Industries). Other fission products are mostly washed to waste. The lanthanides and americium are eluted off the RE-Spec* column, chemically adjusted and evaporated to dryness to remove residual organics and chlorides. The dry sample is dissolved in NH_4SCN -formic acid and passed through a TEVA-Spec* column where the americium is retained (* registered trademark of EIChroM Industries). The solution sample is then passed through a cation exchange column to remove excess NH_4SCN and to separate the lanthanide elements of interest from most of the other lanthanides. Lanthanide recovery is followed through each step by monitoring the $^{154,155}\text{Eu}$ activity.

The U/TEVA-Spec*, RE-Spec*, and associated pre-filter columns are operated in serial configuration with the U/TEVA-Spec* column on top, the pre-filter column in the middle, and the RE-Spec* column on the bottom. All three columns are prewashed with nominally 2 mL of water and 5 mL of 3 M HNO₃. Samples are loaded on the top column, followed by five 1 mL washes of 3 M HNO₃. As stated previously, the U/TEVA-Spec* retains the U and tetravalent actinides. (The resin capacity is about 10 mg of U and can be visually monitored by the yellow color of the absorbed U on the column.) The pre-filter column collects any extractant (resins) which may elute from the U/TEVA-Spec* column. The U/TEVA-Spec* and pre-filter columns are discarded along with all the effluents (washes).

The RE-Spec* column is converted to the chloride form and residual, non-lanthanide fission products are removed by sequentially washing the column with 10 mL of 1 M HNO₃ -0.01 M ammoniumbioxalate, 10 mL of 1 M HNO₃, and 6 mL of 9 M HCl. The effluents are discarded. The lanthanides and Am retained on the RE-Spec* column are eluted from the column with 20 mL of 4 M HCl, then evaporated to dryness and converted to wet ash with HNO₃-H₂O₂ and aqua regia to destroy any traces of organics. Following conversion to HNO₃, the sample is evaporated just to dryness. The sample is dissolved in 1 mL of 2 M NH₄SCN -0.1 M formic acid, covered tightly and allowed to stand to assure the dissolution of the sample.

A TEVA-Spec* column, a pre-filter column and a cation exchange column are operated in series to separate Am from the sample and to obtain a purified lanthanide sample containing the elements of interest (Ce through Gd). The TEVA-Spec* column is prepared by washing the column with 3 mL of water followed by 5 mL of 2 M NH₄SCN -0.1 M formic acid. The pre-filter column is prepared as stated above. The cation exchange column consists of a 2 mL Dowex*50X4 100-200 mesh column that has been washed with 5mL of water followed by 5 mL of 1 M NH₄SCN -0.1 M formic acid (* Dowex is a registered trademark of Dow Chemical Company). The columns are set up in serial configuration with the TEVA-Spec* column on top, the pre-filter column in the middles and the cation exchange column on the bottom. The Am-lanthanide solution sample is transferred to the top column. As in the previous steps, the activity of ^{154,155}Eu is used to follow the Am-lanthanide separation. The columns are washed with 13 mL of 1 M NH₄SCN -0.1 M formic acid and discarded along with all the effluents (the TEVA-Spec* column retains the Am). The Dowex*50X4 column washed with 10 mL of 0.4 M HCl and the effluent is discarded. The lanthanides are eluted from the column with 10 mL of 6 M HCl. The solution is evaporated to dryness, wet ash with HNO₃ and then heated gently over a burner flame to drive off volatile salts. The isotopic composition of the purified sample is then determined by standard inductively coupled plasma mass spectrometry measurements.

APPENDIX B

Estimated Uncertainties in Spent Fuel Sample Analyses

APPENDIX B

Estimated Uncertainties in Spent Fuel Sample Analyses

Uncertainties are given in Section 2.0, Radiochemical Procedures, for all of the analyses except for the lanthanide elements. The uncertainties given in Section 2.0 are for relatively common procedures which have had sufficient use and comparison against standards to permit estimating the uncertainties associated with the respective analysis. However, the analytical procedure used to analyze the spent fuel samples for lanthanide nuclides was developed specifically for the spent fuel assays presented in this report. As indicated in Appendix A, recovery of the lanthanide component in spent fuel can be followed closely by monitoring the ^{154}Eu , ^{155}Eu activity. To further reduce the dependency of the results on total lanthanide recovery, the isotopic compositions are determined by mass spectrometry relative to ^{143}Nd and ^{145}Nd for which absolute values existed. Therefore, it was concluded that errors associated with complete lanthanide recovery were negligibly small compared to those incurred in analyzing the mass spectrometry measurements. To assess the reproducibility of the mass spectrometry data portions of the lanthanide component of spent fuel sample 87-81 were analyzed by two laboratories other than the Pacific Northwest Laboratory (PNL). They were the Los Alamos National Laboratory (LANL), Los Alamos, New Mexico and the Khlopin Radium Institute (KRI), St. Petersburg, Russia. Both PNL and LANL analyzed the lanthanide sample by Inductively Coupled Plasma-Mass Spectrometry (ICP-MS). The KRI employed several techniques in analyzing the lanthanide sample including Secondary Ion Mass Spectrometry (SIMS). However, the KRI suggested that the SIMS data be considered indicative of methods application, and that the Sm and Gd results should be doubted (see Appendix E). Consequently, the SIMS data were not considered in comparing the measurement results obtained by the three laboratories. A copy of the LANL and PNL measurement data, as well as the KRI report, is included in Appendices C, D, and E, respectively.

Basically, the approach taken by KRI in analyzing the sample provided by PNL was to chemically separate the lanthanides into individual elements and then with the use of spikes and laser luminescence perform mass spectrometry measurements to obtain absolute quantity values for each of the lanthanide isotopes. All but about 30 ng of the sample sent to the KRI was dissolved in 0.1M nitric acid and divided between three aliquots. Laser luminescence measurements were performed on one of these solutions sample (0.19735g of solution) and compared to measurements on standard solutions containing known amount of Sm and Eu to obtain absolute quantities for Sm and Eu in the sample. These absolute quantities were multiplied by the isotopic abundance figures obtained later from mass spectrometry measurements to obtain the isotopic values.

One of the solution samples (1.6490g of solution) was spiked with known quantities of Nd-142 and Gd-156. After successive coprecipitation on fluoride and hydroxide of lanthanum and dissolution in 0.001M EDTA, the solution was separated into pure fractions of individual elements by electrophoresis separation on to Whatman-31 paper. Zones were identified on the paper strip by gamma measurements and sample were prepared for mass spectrometry measurements to obtain isotopic abundance figures for Nd and Gd (as well as the other lanthanides). These were then used with known (spiked) values for Nd-142 and Gd-156 to obtain the isotopic values for Nd and Gd.

The third aliquot sample (unspiked) was electrophoretically separated into pure fractions also. Mass spectrometry measurements were performed on the zones as in the spike sample above to obtain isotopic abundance values for the lanthanide elements. These abundance figures were used with the laser luminescence quantitative values for Sm and Eu to obtain the isotopic values for Sm and Eu. About 28.56 ng of sample was removed from the sample shipped and prepared for Secondary Ion Mass Spectrometry (SIMS) measurements. As indicated previously, the SIMS data are suspect and are not considered in the data comparisons that follow.

Since the PNL and LANL results are based on mass spectrometry measurements of the total lanthanide sample, these data are in terms of mass numbers relative to an arbitrarily selected mass number. The KRI data are in terms of isotope concentrations because they separated the lanthanide sample into individual elements prior to mass spectrometry measurements. To compare the data and arrive at estimated error limits, the KRI isotopic values having a common mass number have been combined to obtain a value for that mass number (see Appendix C for the sample isotopic concentrations).

As indicated in Section 2 of the report, the neodymium isotopes were determined by an approved ANSI/ASTM standard measurement method (ANSI/ASTM Standard Test Method E321-79). Since this procedure is a nationally accepted method of analyses, the neodymium values reported in Section 3 of the report are considered the most reliable of the lanthanides. Consequently, the data from each of the three laboratories are compared relative to ^{143}Nd and to ^{145}Nd which are considered to be the better neodymium data. The measurement results obtained from the three laboratories are summarized in the following two tables for comparison purposes relative to ^{143}Nd and to ^{145}Nd .

Table B.1 Comparison of Sample 87-81 Lanthanide Analyses Relative to ^{143}Nd

Mass Number	M-XXX Relative to M-143			Average $\pm 1\sigma$
	PNL	KRI	LANL	
143	1.000	1.000	1.000	1.000-
144	1.54	1.54	1.54	1.54 $\pm$ 0.0
145	0.826	0.821	0.820	0.822 $\pm$ 0.003
146	0.801	0.799	0.795	0.798 $\pm$ 0.003
147	0.352	0.327	0.352	0.344 $\pm$ 0.014
148	0.588	0.571	0.574	0.578 $\pm$ 0.009
149	0.0046	0.0038	0.0032	0.0037 $\pm$ 0.0007
150	0.517	0.500	0.516	0.511 $\pm$ 0.009
151	0.014	0.013	0.014	0.014 $\pm$ 0.001
152	0.134	0.129	0.134	0.132 $\pm$ 0.003
153	0.121	0.121	0.124	0.122 $\pm$ 0.002
154	0.063	0.071	0.063	0.066 $\pm$ 0.005
155	0.0068	0.0103	0.0295 ^(a)	0.0086 $\pm$ 0.0025
156	0.070	0.091	0.098	0.086 $\pm$ 0.014
157	0.0007	0.0007	-	0.0007 $\pm$ 0.0
158	0.0145	0.0198	-	0.0172 $\pm$ 0.0037
159	0.0042	-	-	-
160	0.0003	0.0021	-	0.0012 $\pm$ 0.0013

a) Not included in average.

Table B.2 Comparison of Sample 87-81 Lanthanide Analyses Relative to ^{143}Nd

Mass Number	M-XXX Relative to M-143			Average $\pm 1\sigma$
	PNL	KRI	LANL	
143	1.210	1.218	1.220	1.216 $\pm$ 0.005
144	1.859	1.882	1.880	1.874 $\pm$ 0.013
145	1.000	1.000	1.000	1.000-
146	0.970	0.974	0.970	0.971 $\pm$ 0.002
147	0.426	0.398	0.429	0.418 $\pm$ 0.017
148	0.711	0.696	0.700	0.702 $\pm$ 0.008
149	0.0056	0.0047	0.0039	0.0047 $\pm$ 0.0008
150	0.625	0.609	0.630	0.621 $\pm$ 0.011
151	0.0175	0.0159	0.0165	0.0166 $\pm$ 0.0008
152	0.162	0.157	0.163	0.161 $\pm$ 0.003
153	0.146	0.147	0.151	0.148 $\pm$ 0.003
154	0.076	0.087	0.077	0.080 $\pm$ 0.006
155	0.0083	0.0126	0.036 ^(a)	0.0104 $\pm$ 0.0030
156	0.085	0.110	0.119	0.105 $\pm$ 0.018
157	0.0009	0.0008	-	0.0085 $\pm$ 0.0001
158	0.0176	0.0241	-	0.0208 $\pm$ 0.0046
159	0.0051	-	-	-
160	0.0004	0.0025	-	0.0014 $\pm$ 0.0015

a) Not included in average.

The results obtained by the three laboratories indicate that analytical results for the lanthanides of interest to nuclear criticality can be expected to be within the following one sigma limits:

M-155~29%

Eu-153~2%

Sm-152~3%

Sm-151~7%

Sm-150~2%

Sm-149~18%

Sm-147~4%

Nd-145<1%

Nd-143<1%

APPENDIX C

Pacific Northwest Laboratories Mass Spectrometry Measurement Data

APPENDIX C

Pacific Northwest Laboratories Mass Spectrometry Measurement Data

The lanthanide component in each of the three spent fuel sample, 87-81, 87-72, and 87-63 were analyzed by Inductively Coupled Plasma-Mass Spectrometry in accordance with PNL-ALO-280, RO, dated 9/26/90. The measurement results obtained are presented in this appendix.

	A	B	C	D	E	F
1		Bierman REE Fission Product Analysis				
2		Oct. 16, 1992				
3		SAMPLE GT-81				
4		amu	MEAN		Corr cps/amu	Norm to 144*
5			cps/amu			
6						
7		139	206369		227820	0.99738
8		140	239096		242912	1.06345
9		141	208638		208628	0.91336
10		142	208869		207376	0.90787
11		143	150830		148694	0.65097
12		144	233377		228420	1.00000
13		145	126473		122884	0.53797
14		146	122519		119140	0.52158
15		147	53391		52372	0.22928
16		148	88296		87368	0.38249
17		149	698		687	0.00301
18		150	76366		76853	0.33645
19		151	2130		2152	0.00942
20		152	19500		19948	0.08733
21		153	17455		18004	0.07882
22		154	8923		9340	0.04089
23		155	1793		1019	0.00446
24		156	10639		10456	0.04578
25		157	945		110	0.00048
26		158	2783		2159	0.00945
27		159	1166		625	0.00274
28		160	986		48	0.00021
29		161	546		35	0.00015
30		162	542		49	0.00021
31		163	173		-61	-0.00026
32		164	329		-42	-0.00018

ree lps final

SAMPLE 87-81

	A	B	C	D	E	F
33		165	259		303	0.00132
34						
35		*Approximate error: 5%				

amu	87-72 Norm; Ave	87-63 Norm; Ave	Remix La Run 1	Remix Run 1	Oxide Corr Soln	Oxide Corr Factor	BLANK AVE
139	80219	71557	143090	317	665352		15
140	93359	79441	202328	195841	650297		15
141	81184	68755	106249	103519	746498		15
142	81067	69897	132554	132192	277841		15
143	50884	37733	47542	48437	87773		15
144	95528	82549	97782	97307	173131		15
145	45929	36814	33247	32733	61434		15
146	48114	41850	68274	68865	125257		15
147	17634	12926	9451	9651	16		15
148	35999	31049	30630	30716	41954		15
149	222	237	9335	9064	109		15
150	30054	26861	28258	28258	42116		15
151	640	473	4307	4263	18		15
152	6947	5656	17411	17372	28		15
153	7246	6838	4718	4650	32		15
154	3999	3920	15090	14793	54		15
155	893	831	1864	1278	3416	0.00513	15
156	6725	8032	2766	2681	4698	0.00723	15
157	455	389	2185	2035	4648	0.00623	15
158	1536	1677	2847	2972	2132	0.00768	15
159	464	406	208	291	746	0.00850	15
160	520	438	2387	2551	978	0.00565	15
161	265	198	217	175	491	0.00800	15
162	297	243	338	316	670	0.00535	15
163	88	46	46	49	90	0.00600	15
164	163	122	155	122	267	0.00636	15
165	37	28	35	28	74	0.00600	15

amu	Oxide Contrib 87-72	Oxide Contrib 87-63	Oxide Contrib Remix La	Oxide Contrib Remix	cps/ppb	Element	Mass Discr. Factor
139	0	0	0	0	1432	La	1
140	0	0	0	0	1539	Ce	1
141	0	0	0	0	1547	Pr	1
142	0	0	0	0			1
143	0	0	0	0			1
144	0	0	0	0			1
145	0	0	0	0			1
146	0	0	0	0	1575	Nd	1
147	0	0	0	0	1422	Sm	1
148	0	0	0	0			1
149	0	0	0	0			1
150	0	0	0	0			1
151	0	0	0	0			1
152	0	0	0	0			1
153	0	0	0	0	1519	Eu	1
154	0	0	0	0			1
155	412	367	735	2			1
156	675	574	1462	1415			1
157	506	428	662	645	1502	Gd	1
158	622	536	1017	1015			1
159	432	321	404	412	1873	Tb	1
160	539	466	552	549			1
161	367	294	266	262			1
162	258	224	365	369			1
163	106	78	57	58	1574	Dy	1
164	229	197	195	195			1
165	1	1	56	54	1710	Ho	1

amu	Corr cps/amu	Corr cps/amu	Corr cps/amu	Corr cps/amu
	87-72	87-63	Remix La	Remix
139	80204	71542	143075	302
140	93344	79426	202313	195826
141	81169	68740	106234	103504
142	81052	69882	132539	132177
143	50869	37718	47527	48422
144	95513	82534	97767	97292
145	45914	36799	33232	32718
146	48099	41835	68259	68850
147	17619	12911	9436	9636
148	35984	31034	30615	30701
149	207	222	9320	9049
150	30039	26846	28243	28243
151	625	458	4292	4248
152	6932	5641	17396	17357
153	7231	6823	4703	4635
154	3984	3905	15075	14778
155	466	448	1115	1262
156	6035	7443	1289	1251
157	-65	-54	1508	1375
158	899	1126	1814	1943
159	17	70	-211	-136
160	-34	-43	1820	1986
161	-117	-111	-64	-102
162	24	4	-43	-67
163	-33	-46	-25	-24
164	-81	-90	-55	-88
165	20	11	-36	-41

APPENDIX D

Los Alamos National Laboratory Mass Spectrometry Measurement Data

APPENDIX D

Los Alamos National Laboratory Mass Spectrometry Measurement Data

The lanthanide component in spent fuel sample 87-81 was analyzed by Inductively Coupled Plasma-Mass Spectrometry as described in this appendix along with measurement results. The lanthanide sample analyzed by LANL was taken from the larger sample prepared as described in Appendix A and thus was identical to the sample analyzed by PNL (see Appendix C).

Los Alamos

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Dr. Robert Strebin
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March 9, 1993

Dear Bob,

Enclosed are the final results for the rare earth fission product sample. Included is a procedure describing how the samples were run, and several figures which give the raw data and show the calculations. Let me know if you need any other information.

Sincerely,

Deborah J. Figg, Ph.D.
Staff Member
Analytical Chemistry

copies: CLS-1 Group Office
CRM-4, MS A150

PROCEDURE FOR RUNNING RARE EARTH FISSION PRODUCTS

1) Instrument

The instrument used was a VG PlasmaQuad II. Prior to running the rare earth fission product sample all ICP glassware (spray chamber, nebulizer, and torch) were cleaned in aqua regia and all the peristaltic tubing was changed since it was necessary to recycle the sample given its quantity. The instrument was always given at least 30 minutes to warm up. Following this period, the resolution and mass calibration were checked and the ion optics were tuned for maximum signal at $m/z = 153$ (Eu) using a 100 ppb solution of natural abundance La, Ce, Pr, Nd, Sm, and Eu. The typical ion optic settings (in potentiometer turns) were as follows.

Extractor: 1.0	L1: 6.25	L3: 5.99	Front plate: 2.88
Collector: 6.0	L2: 3.12	L4: 7.50	Pole bias: 5.75

The counts for La^{2+} , La^+ , and LaO^+ were measured in order to determine the percentage of La^{2+} to La^+ and LaO^+ to La^+ . The nebulizer gas flow was adjusted so that these percentages were 2% or less. The gas flow rates (L/min.) were as follows.

Coolant: 16	Auxiliary: 0.60	Nebulizer: 0.731
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The incident plasma rf power was 1.3 kW with a reflected power of less than 5 W. The average signal was 1.4×10^6 cps/ppm Eu.

2) Dissolution of Sample

A 1 μg sample was provided to us by Bob Strubing of PNL. We did not confirm the weight of the sample here. 0.250 ml of concentrated HNO_3 was added to the sample vial, causing an immediate reaction with the sample. The acid was swished around the vial and then the contents were poured into a larger vial. Another 0.250 ml aliquot of concentrated HNO_3 was added to the original sample vial, swished around and then poured into the larger vial. The original sample vial was then rinsed into the larger vial several times with 2% HNO_3 . The volume of the larger vial was taken to 25.0 ml. This was the final solution on which the measurements were made.

3) ICP-MS Measurements

The scan parameters used for all sample, blank, and standard runs were as follows.

of channels = 1024
scans = 600
dwell time = 640 μ s

scan range = 134.9 - 176.5 amu

The sample was analyzed 1 time on day 1, 3 times on day 2, and 3 times on day 3. The procedure for each day was as follows, blank, standard, rare earth sample, blank. When the sample was analyzed 3 times in one day the 3 analyses were performed using a single procedure, that is, 3 x blank, 3 x standard, 3 x rare earth sample, 3 x blank. The blanks that were run first in a procedure were used for blank subtraction (see figure 1 for blank data). The last blanks in the procedure were not used in the calculations, instead only for a qualitative check. The standard, ran second in the procedure, was a 10 ppb solution containing natural abundant La, Ce, Pr, Nd, Sm, and Eu. After all the data acquisition it was decided to rerun the standard at 100 ppb in order to obtain a more precise mass bias measurement. The 100 ppb standard was run 6 times, 3 times on 2 different days. See figure 2 for standard data.

Between different sample types 2% HNO₃ solution was rinsed through the instrument until the counts at Eu 153 were less than 50 cps, it was then allowed to rinse for 2 additional minutes. After placing the sipper in a solution, a 2 minute uptake time was allowed prior to data acquisition.

Since the rare earth sample was recycled during analysis, care was taken so as to minimize dilution from the 2% HNO₃ rinse solution. Before and after the sample was introduced to the instrument, air was allowed to run through the tubing and instrument until all the rinse solution had drained into the waste container.

Blank: 2% HNO₃

Standard: 100 ppb solution of natural abundance La, Ce, Pr, Nd, Sm, and Eu in 2% HNO₃.

4) Results

a) Uncorrected Results

The counts obtained for the blank were subtracted from the counts obtained for the rare earth sample. Then the net counts for each isotope was ratioed to that of $m/z = 145$. See figure 3 for the uncorrected ratios and figure 4 for net sample counts.

b) Corrected Results

The data was corrected for the instrumental mass bias that was determined using the natural abundant rare earth 100 ppb standard.

Mass Bias Determination - (See figure 5 for calculations) The counts obtained for the blank were subtracted from the counts obtained for the standard. Then the counts from the standard were normalized based on the percent abundance at each mass. The normalized counts were then fit to a second order polynomial where $x = m/z$ and $y = \text{normalized counts}$. Using that equation, new normalized counts were extracted for each m/z and then ratioed to that of $m/z = 145$. The resulting ratios represent the mass bias at each m/z relative to $m/z = 145$. There is a 12% mass bias over the mass range of 136 to 154. The mass bias for 155 and 156, relative to 145, is assumed to equal 1.02.

Mass Bias Data Correction - (See figure 4 for calculations) The counts obtained for the blank were subtracted from the counts obtained for the rare earth sample. The net counts were then corrected for the mass bias by dividing the counts at each m/z by the mass bias determined for that m/z . These corrected counts were then ratioed to that of $m/z = 145$. The statistics for the corrected ratios are shown in figure 6.

Figure 7 lists the final uncorrected and corrected ratios and their corresponding error. The corrected ratios should be more accurate.

Figure 1: Blank Counts (cps)

m/z	136	137	138	139	140	141	142	143	144	145	146	147	148	149	150
run #1	75	50	197	40	38	36	33	28	30	28	29	28	31	29	30
run #2	35	32	59	32	29	32	31	29	28	31	28	32	32	34	31
run #3	36	33	56	31	31	33	29	32	30	32	28	29	29	29	31
run #4	35	27	53	32	29	29	29	29	29	30	32	32	34	30	30
run #5	63	47	162	30	30	30	30	31	31	30	33	30	29	39	29
run #6	60	48	146	34	41	32	30	32	26	28	26	33	29	30	32
run #7	57	47	133	37	34	32	31	30	30	33	29	32	30	33	34
m/z	151	152	153	154	155	156	157	158	159	160	161	162	163	164	166
run #1	36	29	33	30	28	24	30	29	30	25	28	29	27	29	27
run #2	31	28	29	30	29	27	29	30	30	28	29	28	29	28	28
run #3	27	29	32	31	31	34	31	29	30	34	30	32	30	32	29
run #4	30	31	28	32	35	31	29	32	27	31	27	29	32	30	27
run #5	28	32	31	28	31	29	32	27	31	31	30	26	28	29	30
run #6	30	28	31	30	30	32	29	31	30	29	31	29	33	30	30
run #7	34	30	28	30	29	31	28	31	27	33	29	29	31	29	29

Figure 2: Data for Natural Abundant Standard (cps)

m/z	136	138	139	140	141	143	145
run #1	469	804	231435	219453	264218	32226	22479
run #2	461	818	234337	222374	267181	32440	22523
run #3	484	824	242518	229459	274939	33264	23156
run #4	502	919	249812	236979	284679	34570	24044
run #5	508	932	249676	236614	284535	34697	24000
run #6	508	962	251352	238555	288036	35139	24471
average	489	877	243188	230572	277265	33723	23446
std. dev	20.49	68.75	8597.03	8165.31	10011.56	1246.51	846.91
m/z	146	147	149	151	152	153	154
run #1	46958	39185	36888	131559	73090	146802	63097
run #2	46956	39388	36923	132273	73121	147049	63016
run #3	48331	40600	37981	135427	74896	150717	64777
run #4	50451	42280	39632	140559	78017	156434	67088
run #5	50437	42399	39558	140972	77934	156343	67063
run #6	51052	42728	40068	142909	79012	158630	68071
average	49031	41097	38508	137283	76012	152663	65519
std. dev	1853.14	1586.08	1429.25	4843.59	2640.29	5157.97	2192.72

Figure 3: Statistics for Uncorrected Ratios

ratio	136/145	137/145	138/145	139/145	140/145	141/145	142/145	143/145
run #1	0.00565	0.00758	0.04987	1.68914	1.92111	1.65624	1.67428	1.20697
run #2	0.00492	0.00671	0.04357	1.48644	1.76736	1.57488	1.56516	1.18703
run #3	0.00474	0.00675	0.04453	1.49578	1.77055	1.57617	1.56759	1.18713
run #4	0.00483	0.00665	0.04437	1.50142	1.77584	1.57833	1.56117	1.19022
run #5	0.00576	0.00801	0.05160	1.69994	1.94227	1.67432	1.68623	1.21045
run #6	0.00581	0.00796	0.05149	1.69695	1.92972	1.66292	1.68261	1.20432
run #7	0.00576	0.00798	0.05132	1.70964	1.94692	1.67520	1.68893	1.21178
average	0.00535	0.00738	0.04811	1.61133	1.86482	1.62829	1.63228	1.19970
std. dev.	0.00049	0.00065	0.00375	0.10949	0.08796	0.04893	0.06346	0.01113
ratio	144/145	146/145	147/145	148/145	149/145	150/145	151/145	152/145
run #1	1.87479	0.97141	0.42571	0.70470	0.00380	0.62029	0.01619	0.15968
run #2	1.85909	0.98256	0.44357	0.73017	0.00418	0.66040	0.01737	0.17431
run #3	1.85601	0.97679	0.44243	0.72584	0.00391	0.65253	0.01730	0.17347
run #4	1.86218	0.97731	0.44210	0.72796	0.00416	0.66082	0.01783	0.17409
run #5	1.88168	0.97789	0.42916	0.70818	0.00386	0.62787	0.01650	0.16180
run #6	1.87453	0.97127	0.42539	0.70609	0.00384	0.62412	0.01657	0.16189
run #7	1.87978	0.97373	0.42874	0.70868	0.00403	0.62531	0.01645	0.16219
average	1.86972	0.97585	0.43387	0.71595	0.00397	0.63876	0.01689	0.16678
std. dev.	0.01042	0.00403	0.00839	0.01141	0.00016	0.01825	0.00061	0.00677
ratio	153/145	154/145	155/145	156/145				
run #1	0.14569	0.07385	0.03988	0.11768				
run #2	0.16215	0.08258	0.03855	0.12575				
run #3	0.16100	0.08188	0.04065	0.12724				
run #4	0.16148	0.08264	0.04026	0.12744				
run #5	0.14930	0.07567	0.03158	0.11527				
run #6	0.14791	0.07583	0.03246	0.11634				
run #7	0.14947	0.07613	0.03452	0.11943				
average	0.15386	0.07837	0.03684	0.12131				
std. dev.	0.00730	0.00382	0.00389	0.00533				

[illegible]

Figure 4: data correction

corrected counts (cps) run #4	ratio to 145	blank subtracted counts (cps) run #5	corrected counts (cps) run #5	ratio to 145	blank subtracted counts (cps) run #6	corrected counts (cps) run #6	ratio to 145	blank subtracted counts (cps) run #7	corrected counts (cps) run #7	ratio to 145	m/z
609.62073	0.00532	765	841.804799	0.00634	732	805.491651	0.00640	695	764.776909	0.00633	136
826.087986	0.00721	1063	1152.40358	0.00868	1002	1086.27318	0.00863	964	1045.07719	0.00865	137
5437.35172	0.04745	6851	7325.72206	0.05517	6484	6933.29176	0.05506	6197	6626.40484	0.05487	138
181674.338	1.58529	225725	238333.866	1.79490	213682	225618.151	1.79174	206454	217986.399	1.80514	139
212408.026	1.85347	257902	268176.874	2.02718	242992	253615.043	2.01408	235108	245386.373	2.03203	140
186807.948	1.63009	222323	229612.96	1.72922	209396	216262.084	1.71744	202295	208928.243	1.73013	141
183026.535	1.59709	223905	229056.823	1.72503	211876	216751.048	1.72133	203954	208646.771	1.72779	142
138351.719	1.20726	160728	163029.019	1.22778	151649	153820.042	1.22156	146333	148427.937	1.22913	143
214827.516	1.87459	249857	251521.319	1.89421	236043	237615.303	1.88702	227000	228512.067	1.89230	144
114600	1.00000	132784	132784	1.00000	125921	125921	1.00000	120759	120759	1.00000	145
111361.136	0.97174	129848	129107.328	0.97231	122303	121605.366	0.96573	117587	116916.267	0.96818	146
50133.9018	0.43747	56986	56388.6416	0.42466	53565	53003.5024	0.42093	51774	51231.2767	0.42424	147
82225.9534	0.71750	94035	92684.5695	0.69801	88912	87635.1406	0.69595	85580	84350.9912	0.69851	148
468.719316	0.00409	513	504.094359	0.00380	484	475.597797	0.00378	487	478.545717	0.00396	149
74253.6218	0.64784	83371	81745.6583	0.61563	78590	77057.8653	0.61195	75512	74039.8718	0.61312	150
2000.5502	0.01746	2191	2145.47503	0.01616	2087	2043.63596	0.01623	1987	1945.71377	0.01611	151
19527.7349	0.17040	21485	21029.1907	0.15837	20385	19952.5275	0.15845	19586	19170.4785	0.15875	152
18120.9135	0.15812	19825	19412.4668	0.14620	18625	18237.4373	0.14483	18050	17674.4023	0.14636	153
9284.79922	0.08102	10048	9851.49552	0.07419	9548	9361.27381	0.07434	9193	9013.2164	0.07464	154
4523.52941	0.03947	4193	4110.78431	0.03096	4087	4006.86275	0.03182	4169	4087.2549	0.03385	155
14318.6275	0.12494	15306	15005.8824	0.11301	14650	14362.7451	0.11406	14422	14139.2157	0.11709	156

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Figure 5: Mass Bias Determination

				x=mass	
				normalized counts	mass bias
	natural abundance	counts from nat.	normalized	as extracted from	relative to
mass	at that mass (%)	abund. std. (cps)	counts (cps)	2nd order fit	m/z=145
136	0.19	489	257368.4211	256348.928	0.90876
137				260201.702	0.92242
138	0.09+0.26=0.35	876	250285.7143	263806.2	0.93520
139	99.9	243108	243431.4314	267162.422	0.94710
140	88.5	230572	260533.3333	270270.368	0.95811
141	100	277265	277265	273130.038	0.96825
142	11.1+27.1=38.2	104397	273290.5759	275741.432	0.97751
143	12.2	33723	276418.0328	278104.55	0.98589
144	23.9+3.1=27.0	74939	277551.8519	280219.392	0.99338
145	8.3	23445	282469.8795	282085.958	1.00000
146	17.2	49031	285063.9535	283704.248	1.00574
147	15	41097	273980	285074.262	1.01059
148	5.7+11.2=16.9	47969	283840.2367	286196	1.01457
149	13.8	38508	279043.4783	287069.462	1.01767
150	5.6+7.4=13.0	37498	288446.1538	287694.648	1.01988
151	47.8	137283	287202.9289	288071.558	1.02122
152	26.7	76012	284689.1386	288200.192	1.02168
153	52.2	152662	292455.9387	288080.55	1.02125
154	22.8	65519	287364.0351	287712.632	1.01995
155					1.02000
156					1.02000
157					
158					
159	The normalized counts were fit to a 2nd order polynomial				
160	$y = -26565.49 + 38239x - 124.138x^2$ $R=0.883$				
161					

Figure 6: Statistics for Corrected Ratios

ratio	136/145	137/145	138/145	139/145	140/145	141/145	142/145	143/145
run #1	0.00621	0.00822	0.05332	1.78350	2.00510	1.71054	1.71280	1.22425
run #2	0.00542	0.00727	0.04659	1.56947	1.84462	1.62652	1.60117	1.20403
run #3	0.00522	0.00732	0.04762	1.57933	1.84795	1.62786	1.60366	1.20412
run #4	0.00532	0.00721	0.04745	1.58529	1.85347	1.63009	1.59709	1.20726
run #5	0.00634	0.00868	0.05517	1.79490	2.02718	1.72922	1.72503	1.22778
run #6	0.00640	0.00863	0.05506	1.79174	2.01408	1.71744	1.72133	1.22156
run #7	0.00633	0.00865	0.05487	1.80514	2.03203	1.73013	1.72779	1.22913
average	0.00589	0.00800	0.05144	1.70134	1.94635	1.68169	1.66984	1.21687
std. dev.	0.00054	0.00070	0.00401	0.11561	0.09181	0.05053	0.06492	0.01129
ratio	144/145	146/145	147/145	148/145	149/145	150/145	151/145	152/145
run #1	1.88728	0.96587	0.42124	0.69458	0.00373	0.60820	0.01585	0.15629
run #2	1.87147	0.97695	0.43892	0.71968	0.00411	0.64752	0.01701	0.17061
run #3	1.86837	0.97121	0.43779	0.71541	0.00384	0.63981	0.01694	0.16979
run #4	1.87459	0.97174	0.43747	0.71750	0.00409	0.64794	0.01746	0.17040
run #5	1.89421	0.97231	0.42466	0.69801	0.00380	0.61563	0.01616	0.15837
run #6	1.88702	0.96573	0.42093	0.69595	0.00378	0.61195	0.01623	0.15845
run #7	1.89230	0.96818	0.42424	0.69851	0.00396	0.61312	0.01611	0.15875
average	1.88218	0.97028	0.42932	0.70566	0.00390	0.62631	0.01654	0.16324
std. dev.	0.01049	0.00401	0.00830	0.01124	0.00015	0.01790	0.00059	0.00663
ratio	153/145	154/145	155/145	156/145				
run #1	0.14266	0.07241	0.03910	0.11537				
run #2	0.15878	0.08097	0.03779	0.12329				
run #3	0.15765	0.08028	0.03985	0.12474				
run #4	0.15812	0.08102	0.03947	0.12494				
run #5	0.14620	0.07419	0.03096	0.11301				
run #6	0.14483	0.07434	0.03182	0.11406				
run #7	0.14636	0.07464	0.03385	0.11709				
average	0.15066	0.07684	0.03612	0.11893				
std. dev.	0.00715	0.00374	0.00381	0.00522				

Figure 7: Final Results

	uncorrected	corrected
	ratio to	ratio to
isotope	m/z=145	m/z=145
136	0.0054 $\pm$ 0.0005	0.0059 $\pm$ 0.0005
137	0.0074 $\pm$ 0.0006	0.0080 $\pm$ 0.0007
138	0.048 $\pm$ 0.004	0.051 $\pm$ 0.004
139	1.6 $\pm$ 0.1	1.7 $\pm$ 0.1
140	1.86 $\pm$ 0.09	1.95 $\pm$ 0.09
141	1.63 $\pm$ 0.05	1.68 $\pm$ 0.05
142	1.63 $\pm$ 0.06	1.67 $\pm$ 0.06
143	1.20 $\pm$ 0.01	1.22 $\pm$ 0.01
144	1.87 $\pm$ 0.01	1.88 $\pm$ 0.01
145	1	1
146	0.976 $\pm$ 0.004	0.970 $\pm$ 0.004
147	0.434 $\pm$ 0.008	0.429 $\pm$ 0.008
148	0.72 $\pm$ 0.01	0.70 $\pm$ 0.01
149	0.0040 $\pm$ 0.0002	0.0039 $\pm$ 0.0002
150	0.64 $\pm$ 0.02	0.63 $\pm$ 0.02
151	0.0169 $\pm$ 0.0006	0.0165 $\pm$ 0.0006
152	0.167 $\pm$ 0.007	0.163 $\pm$ 0.007
153	0.154 $\pm$ 0.007	0.151 $\pm$ 0.007
154	0.078 $\pm$ 0.004	0.077 $\pm$ 0.004
155	0.037 $\pm$ 0.004	0.036 $\pm$ 0.004
156	0.121 $\pm$ 0.005	0.119 $\pm$ 0.005
157	0.027 $\pm$ 0.002	
158	0.048 $\pm$ 0.002	
159	0.019 $\pm$ 0.001	
160	0.026 $\pm$ 0.002	
161	0.014 $\pm$ 0.001	

APPENDIX E

Khlopin Radium Institute Measurement Data

APPENDIX E

Khlopin Radium Institute Measurement Data

The Khlopin Radium Institute in St. Petersburg, Russia, employed several measurement approaches to analyze the lanthanide component in spent fuel sample 87-81. The lanthanide sample analyzed by the KRI was taken from the larger sample prepared as described in Appendix A and thus was identical to the sample analyzed by PNL and LANL (see Appendices C and D). The measurement techniques used by the KRI were briefly discussed in Appendix B in connection with estimating uncertainties associated with the lanthanide concentrations in the spent fuel samples. More detailed descriptions of the measurements and additional data are presented in the KRI report on the analyses of sample 87-81. The report is reproduced in this appendix.

European Analytical Services, Inc.
(Representing the V.G.Khlopin Radium Institute,
St.Petersburg, Russia)

A.A.Rimski-Korsakov, A.V.Stepanov, A.D.Kirikov

RADIOCHEMICAL ANALYSIS OF SPENT REACTOR FUEL SAMPLE

Report of Results

Work performed on :

Contract on Purchase Order No. 190116-A-M2, of February 1993
for Batelle Pacific Northwest Laboratories, 5-th & W Streets,
P.O.Box 999, Richland, Washington 99352, USA

St.Petersburg

1993

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C O N T E N T S

1. General Scope of the Problem and Statement of Work
2. Sample Description
3. Analytical Methods Applied - Short Description
4. Standards Used in the Analysis
5. Analytical Results

1. General Scope of the Problem and Statement of Work

A sample of rare-earth fraction has been obtained from irradiated reactor fuel and presented for analysis of abundance of several nuclides :

Sm - 147, 149, 150, 151, 152

Gd - 155

Eu - 155

Nd - 143, 145

Concentrations were to be measured either in grams of nuclide per sample or as the amount of nuclide relative to Nd-145.

Among the requirements of the Statement of Work a review of results by a technically qualified individual other than the analyst was stated.

2. Initial Sample Evaluation and Preparation

The sample was in the form of several milligrams of dry residue in a glass vial, packed in lead container and in the shipping drum. This package has been transported by Air Freight to St. Petersburg and received from customs in May 1993 - undamaged and sealed.

The HPGe gamma-spectrometer was used to measure the activities in the sample. The only two nuclides detected were 154-Eu and 155-Eu :

154-Eu 840 +/- 100 KBq i.e. 80 +/- 10 ng

155-Eu 230 +/- 30 Kbc i.e. 13 +/- 2 ng

Procedure used complied with the OST 95 10234-86(1.11.1987) Standard - and the sample had not to be removed from the original vial. This measurement was made only to evaluate the expected amounts of the fission products (both radioactive and stable) present in the sample and will be compared later with amounts detected by mass-spectrometry.

Three different methods were chosen for the analysis of the sample : the secondary ion mass spectrometry (SIMS) for panoramic investigations of isotopic ratios of rare earths, laser induced fluorimetry for direct determination of total Eu and Sm content in the sample and conventional isotope dilution mass spectrometry with preliminary radiochemical isolation of pure Nd, Sm, Eu and Gd for precise determination of isotope abundances of those elements and total amounts of Nd and Gd.

Two of these methods (SIMS and fluorimetry) can use the microgram quantity of material, while the latter needed as much of the sample material as possible.

For the first method (SIMS) a small (about 1/1000) aliquot was taken from the sample. This was done by touching the inner wall of the sample vial with the end of a clean platinum wire slightly wetted with 0.1 M nitric acid and letting it dry. Repeated gamma-spectrometry measurement has shown that about 600 Bq of ^{154}Eu was present on the platinum wire, and consequently less than 1/1000 of the original sample material has been removed. That platinum wire was used in the SIMS analysis as described below, and the rest of the sample went to the radiochemical analysis. Since all errors in absolute isotope concentrations in the sample weights are not less than several %, we did not correct the results for this (negligibly small) part of the sample we removed.

3. Analytical Methods Applied - Short Description

3.1. SIMS analysis procedure and corrections

Secondary Ion Mass Spectrometry (SIMS) has been performed with a very small portion of the sample (after conventional radiochemical determination the whole PEE content of sample was found to be about 40 milligrams and the SIMS part of the sample was determined to have about 30 nanograms, derived as shown in section 2.). The platinum wire with small part of the original sample on it's surface has been exposed to the 10 nA beam of negative $^{16}\text{O}^-$ ions in IMS 4f Ion Microprobe (CAMECA) mass-spectrometer. In order to reduce the molecular ion interference the energy-filtering technique has been applied using sample HV offset (-100 V) and small energy slit (10 eV). Nevertheless the difference in the evaporation efficiency for different elements (Nd, Sm, Eu and Gd) had to be corrected for using the standard sample with known concentrations of the same elements. This standard had been prepared from natural isotope mixtures of these elements and certified by the State Institute "Mekhanobr" in St. Petersburg (see Section 4, below). Correction factors (i.e. ratios of true element concentration to one derived from the mass-spectrum) found this way were :

$$C(\text{Nd}) = 1. ; C(\text{Sm}) = 0.91 ; C(\text{Eu}) = 0.88 ; C(\text{Gd}) = 0.94$$

The total mass-spectrum received from IMS 4f spectrometer gave mass abundances for all (cumulative) masses from 142 to 160. Those masses that were undoubtedly belonging to single isotope components of definite elements (masses 142 - 147, 149, 153, 156 - 160) were corrected using the coefficients quoted above. Those mass peaks that in all probability belonged to several (mostly two) isotopes of different elements were corrected using the average of the corresponding coefficients (and the relative error quoted was increased by 17% - the typical difference in the coefficients). Later the masses for such doublets are quoted in the results as sums of all element components for this particular mass value. Final results obtained by SIMS method are presented in Section 5, below.

3.2. Laser luminescence determination of Sm and Eu

Absolute measurement of Sm and Eu amount in the sample was done by luminescent analysis. Luminescence was induced by UV light from a nitrogen laser in the sensibilizing solution of tenciltrifluoracetone + TOPO. The measurement was done relatively to luminescence of standard solutions containing known quantities of Sm and Eu. Preparation of the standards and their characteristics are presented in Section 4, below. Standards were prepared using natural isotopic mixtures of Sm and Eu, containing 56.90 ng/g of Sm in standard solution and 5.34 ng/g of Eu in standard solution. Concentrations in the sample were derived from the ratios between number of PMT counts for equal volumes of solution analysed and the standard. These results gave absolute (ng/sample) values for the most "difficult" pair Sm - Eu, which were later just multiplied by the isotope abundance figures derived from the mass-spectrometry data.

3.3. Radiochemical separation / mass-spectrometry procedure

The dried sample was dissolved in concentrated nitric acid, evaporated to dryness and dissolved in 0.1 M nitric acid. Weight of the empty vial - 19.2938 g, with solution - 22.3936 g, solution weight - 3.0998 g. An aliquot of 0.19735 g was taken from that solution for laser luminescence analysis of Eu and Sm (see in section 3.2.) Remaining solution has been divided into two parts.

The first part weighting 1.5490 g was used for preparation of

isotopically spiked solution and quantitative analysis of Nd and Gd in the sample. For this a known quantity of isotope spikes Nd-142 and Gd-156 were added into this part. The second part was used for isotope assay determinations. Rare earths in both parts were purified by means of successive coprecipitation on fluoride and hydroxide of lanthanum of high purity. After dissolution in 0.001 M EDTA having $pH = 1.7$ the solution was separated by electrophoresis on Whatman-31 paper into pure fractions of individual elements (for subsequent mass-spectrometric investigation). Conditions of electrophoretic separation: $E=24$ V/cm ; $t=70$ minutes.

Zone positions on the paper strip were located by scanning it with a collimated GM-counter. As an example the electrophoregram for unspiked part of the sample is given on Fig. 1

The following parts of the paper strip were cut off and used for the mass-spectrometric measurement of the isotopic composition of each element (from the start) :

Nd - 29-30 cm, Sm - 16-16.5 cm, Eu - 13.5-14 cm, Gd - 10.5-11 cm

These sections were ashed, dissolved and deposited on the tantalum filaments for TE-6 mass-spectrometer.

The results of mass distribution determinations for Nd, Sm, Eu and Gd fractions are presented in Section 5, below. For calculation of Nd and Gd amount the isotopic ratios relative to Nd-144 and Gd-156 were measured in unspiked and spiked parts of the sample. For calculation of element concentrations the well known equation of isotope dilution method was used. Results of determinations are given below in Tables 5.1-5.2.

4. Standards used in the analysis =

The calibration of laser fluorimeter and IDMS were made with use of working reference solutions prepared from pure oxides of Nd, Sm, Eu and Gd in accordance with russian standard GOST 23262.0-79. According to information obtained from producer all four oxides were of 99.97, 99.75, 99.94 and 99.9996 % purity respectively. After calcination till the constant weight was achieved all oxides were dissolved in nitric acid and final solutions were weighed. Those stock solutions were used for preparation of working reference solutions.

Weights of oxides of four elements taken for preparation of stock solutions, their concentrations and the concentrations of reference solutions used for calibration of analytical methods are given below :

	g	mg/g	mcg/g
Nd	0.35004	9.0171	
Sm	0.21341	6.0146	9.4316
Eu	0.34469	9.2969	1.9154
Gd	0.23001	4.9476	1.0803

The content of main isotopes in isotopic tracers Nd-142 and Gd-156 was 95.7 and 95.5 wt % respectively. Their concentrations in spike solutions determined by means of isotope dilution mass spectrometry with use of working reference solutions of Nd and Gd of natural isotopic composition as tracers were as follows:

Nd-142 - $7.59 \pm 0.025 \mu\text{g/g}$; Gd-156 - $4.80 \pm 0.03 \mu\text{g/g}$;

5. Analytical results

Results of analyses are presented below in tables 5.1., 5.2. and 5.3. The determination of total amount of Sm and Eu by fluorimetry gave the values 8725 ± 100 and 1223 ± 21 ng per sample respectively for $p = 0.95$. Of course the radiochemical separation and mass-spectrometry with isotopic dilution gives much more detailed and reliable data, and permits to determine the absolute nanograms per sample (Tables 5.1. & 5.2.). The SIMS analysis (Table 5.3.) gives only relative (to Nd-145) concentrations and does not resolve the multielement peaks. Nevertheless we think that comparison of two sets of data shows applicability of SIMS to some problems in this field and should be kept in mind when fast analysis of very small quantities is necessary. We suggest that SIMS data for masses involving Sm isotopes should be doubted - probably because the correction factor for Sm in the sample has been erroneous - due to some difference in the chemical properties of the standard and the sample analysed. We marked the doubtful lines with "*".

Table 5.1.

Absolute nanograms per sample measured by radiochemical separation and mass-spectrometry with isotopic dilution method.

Mass number	Nanograms per sample $\pm$ error			
	Neodymium	Samarium	Europium	Gadolinium
142	287 $\pm$ 3			
143	9100 $\pm$ 40			
144	14060 $\pm$ 55			
145	7470 $\pm$ 32			
146	7270 $\pm$ 35	4 $\pm$ 5		
147		2975 $\pm$ 100		
148	3910 $\pm$ 24	1290 $\pm$ 20		
149		35 $\pm$ 5		
150	1850 $\pm$ 150	2596 $\pm$ 110		
151		96 $\pm$ 35	23 $\pm$ 2	
152		1160 $\pm$ 60	11 $\pm$ 10	.3 $\pm$.3
153			1100 $\pm$ 20	
154		393 $\pm$ 20	79 $\pm$ 7	176 $\pm$ 4
155			13 $\pm$ 2	81 $\pm$ 2
156				825 $\pm$ 16
157				6 $\pm$ 4
158				180 $\pm$ 4
159				
160				19 $\pm$ 2

Table 5.2.

Isotope concentrations relative to the ^{145}Nd concentration calculated from measurements by radiochemical separation and mass-spectrometry with isotopic dilution method.

Mass number	Ratio of isotope to ^{145}Nd $\pm$ error			
	Neodymium	Samarium	Europium	Gadolinium
142	0.039 $\pm$ 0.001			
143	1.218 $\pm$ 0.008			
144	1.882 $\pm$ 0.011			
145	1.000			
146	0.973 $\pm$ 0.006			
147		0.398 $\pm$ 0.013		
148	0.523 $\pm$ 0.004	0.173 $\pm$ 0.003		
149		0.005 $\pm$ 0.001		
150	0.248 $\pm$ 0.020	0.361 $\pm$ 0.015		
151		0.013 $\pm$ 0.005	0.0031 $\pm$ 0.0003	
152		0.155 $\pm$ 0.005	0.0002 $\pm$ 0.0002	< 0.00004
153			0.1472 $\pm$ 0.0027	
154		0.053 $\pm$ 0.003	0.0105 $\pm$ 0.0009	0.0236 $\pm$ 0.0005
155			0.0018 $\pm$ 0.0003	0.0108 $\pm$ 0.0004
156				0.110 $\pm$ 0.00 2
157				< 0.00007
158				0.0241 $\pm$ 0.0005
159				
160				0.0025 $\pm$ 0.0003

Table 5.3.

Isotope concentrations relative to the ^{145}Nd concentration
measured by secondary ion mass-spectrometry (SIMS) method

Mass number	Ratio of isotope to ^{145}Nd $\pm$ error			
	Neodymium	Samarium	Europium	Gadolinium
142	0.024 $\pm$ 0.010			
143	1.22 $\pm$ 0.03			
144	1.87 $\pm$ 0.03			
145	1.000			
146	0.96 $\pm$ 0.03			
147 *		0.46 $\pm$ 0.06		
148	(Nd+Sm summed = 0.66 $\pm$ 0.08)			
149 *		0.008 $\pm$ 0.003		
150	(Nd+Sm summed = 0.65 $\pm$ 0.08)			
151 *		(Sm+Eu summed = 0.024 $\pm$ 0.005)		
152 *		(Sm+Eu summed = 0.17 $\pm$ 0.03)		
153		0.16 $\pm$ 0.02		
154		(Sm + Eu + Gd total for 154 = 0.076 $\pm$ 0.06)		
155 *		(Eu+Gd summed = 0.005 $\pm$ 0.003)		
156				0.07 $\pm$ 0.03
157				< 0.002
158				< 0.002
159				
160				< 0.002

Sample USA 07-81, unspiked, sep-n 17-09-1993
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