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Nd and Sm Isotopic Composition of Spent Nuclear Fuels from Three Material Test Reactors

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Abstract

Rare earth elements such as neodymium and samarium are ideal for probing the neutron environment that spent nuclear fuels are exposed to in nuclear reactors. The large number of stable isotopes, some having very large neutron capture cross sections, can provide distinct isotopic signatures for differentiating the source material for nuclear forensic investigations. Measurements of the isotopic abundances were carried out by first chromatographically separating the elements of interest prior to isotopic analysis using multi-collector, inductively coupled plasma, mass spectrometry (MC-ICP-MS). The rare-earth elements were isolated from the high activity fuel matrix via ion exchange chromatography in a shielded cell. The resulting samples then have a low enough activity to be handled outside the glove box or isolated environment, and the individual elements can be separated using cation exchange chromatography with alpha-hydroxyisobutyric acid. The neodymium and samarium aliquots were analyzed via MC-IPC-MS, resulting in isotopic compositions with a precision of 0.01-0.3 %.

Introduction

Two types of high-profile nuclear materials in the field of nuclear forensics are pre-detonation and post-detonation nuclear material. Pre-detonation materials can vary in composition and complexity from uranium ores to spent nuclear material. Spent nuclear material represents a unique security concern as it can be used directly in a radiological dispersion device and it still contains significant amounts of unreacted ^{235}U. Nuclear fuel from material test reactors (MTR), a subclass of research reactors, operated with highly enriched ^{235}U (HEU) fuels, in some cases up to 93%, until the 1970s when in response to growing security concerns the

research reactors were converted to low-enriched fuel (LEU) with enrichments on the order of 20% (1). With their large amount of fissile material and radioactivity, spent nuclear fuels represent high-profile targets for illicit activities. Methods capable of determining an interdicted fuel rod's provenance and host-reactor's operating characteristics are required should spent nuclear fuel provenance ever be questioned (2).

During neutron induced fission of ^{235}U and ^{239}Pu various rare earth elements (e.g., La-Dy) are produced on the high-mass tail of the fission curve. Of these elements, Nd, Sm, and Gd have multiple stable isotopes and are produced in measureable quantities in non-natural isotopic abundances (3). These elements also have isotopes that have large (> 1,000 b) thermal neutron capture cross sections, resulting in one-mass heavier isotopes that have negligible (≤ 1b) capture cross sections. Consequently, depletion occurs in the large capture cross section isotopes and enrichment occurs in the low capture cross section isotopes that are proportional to the total neutron fluence the isotopes have been experienced. Therefore, the absolute and relative isotopic abundances of Nd, Sm, and Gd can provide important information regarding the type of fissile material powering a chain reaction along with details about the neutron environment inside the reactor.

In prior studies Nd, Sm, and Gd isotopic composition in spent nuclear fuel have been analyzed using high-performance liquid chromatography (HPLC) and ion exchange systems coupled to mulit-collector inductively coupled mass spectrometers (MC-ICP-MS) (4-6) to separate the individual elements. However, both methods require dedicated instrumentation at least partially contained in a glovebox to perform the separations, which can be logistically challenging. Other studies (7, 8) have used column chromatography to analyze spent nuclear fuel from pressurized water reactors, which use fuels that are typically enriched to ~2-5% ^{235}U.

This study presents a simplified method for separating Nd and Sm from spent nuclear fuel using a series of cation exchange chromatographic separations optimized for high-dose remote handling situations allowing subsequent chemistry and analyses to be performed outside of high-dose environments. The short-lived radionuclides were removed using chromatographic columns in a shielded cell (9) prior to sample removal. Chromatographic purification of Nd and Sm were conducted in a radiological hood before being analyzed using multi-collector inductively coupled plasma mass spectrometry. Spent fuels and reference materials (BHVO-2G, a powdered basalt) were treated with the same column chemistry, which enabled the reference materials to serve as a control for mass fractionation correction. The Nd and Sm isotopic compositions of three spent fuels are compared to determine if specific reactor characteristics can be distinguished.

Materials and Methods

Fuel Rod Sampling and Lanthanoid Separation

Three MTR spent nuclear fuel assemblies were sampled individually in 2009, 2010, and 2011 (here on referred to as A, B, and C, respectively) at the Savannah River National Laboratory (SRNL) Shielded Cells Facility. Declared initial and spent ^{235}U enrichments for each assembly are listed in Table 1. During each sampling event, the individual fuel assembly was placed into a containment device inside a shielded cell. The containment device ensured that no cross contamination occurred between the shielded cells environments and the fuel assemblies. The spent fuel assemblies from the MTR consisted of multiple plates of uranium fuel spatially separated from one another and encased in aluminum cladding. Five to six core samples were excised evenly down the length of the fuel rod, with only the top fuel plate being sampled. The sampling process provided sufficient mass of material to obtain representative compositional

analyses along the length of the fuel rod while minimizing the radioactivity of the processed solution. The extracted material had an average weight of approximately 1.5 g total with around 0.2 g consisting of the uranium fuel and the rest consisting of the Al-cladding material.

The sampled fuels were each dissolved in 100 mL of 50% (v/v) aqua regia. Procedural blanks were prepared in the containment device for each fuel rod sampling event, resulting in three blanks labeled Blank A, Blank B, and Blank C with letters identifying the containment device from which the fuel rod was sampled. These blank solutions were treated identically to the fuel samples and their concentrations and isotopic abundances are reported along with that of the fuel rods.

All acids (alpha-hydroxyisobutyric, HCl and HNO_3) used in this work were either purchased (99.99% grade Optima, Fisher Scientific, Pittsburgh, PA, U.S.A.) or purified using a triple-distillation process in-house at the University of Maryland (UMD), along with 18 MΩ water, which was used for dilutions and chemistry. Prior to use, the chromatographic resins (AG-50W, x8, 200-400 mesh) were cleaned with 6 M HCl and 18 MΩ water rinses. All sample fractions were collected in acid-leached PTFE containers and standard reference material chemistry was performed in a class-100 clean laboratory in the Department of Geology at UMD.

The lanthanoids were isolated and individually separated similarly to methods developed for the analysis of trinitite and samples from Oklo (10, 11). Lanthanoid separations for samples and blanks A-C were performed in the Shielded Cells Facility at SRNL without the use of a containment device. An aliquot of the 100 mL fuel and blank solutions was transferred to a scintillation vial pre-marked at the 4 mL volume level. These vials were dried down overnight in an oven at 70^{o}C and then diluted with 2 mL of 2.0 M HCl. The lanthanoids were separated from the highly-radioactive fission products (primarily ^{137}Cs and ^{90}Sr and their daughters due to

131 significant time between reactor discharge and analysis), uranium, plutonium, and the aluminum-
132 rich matrix using cleaned cation resin contained in 10 cm x 1.5 cm plastic columns
133 (Environmental Express R1020) and pre-conditioned with 2.0 M HCl. After loading the samples
134 onto the columns, the resin was washed with 60 mL of 2.0 M HCl followed by 15 mL of H_2O
135 and 50 mL of 2.0 M HNO_3. The lanthanoids were eluted and collected with 50 mL of 4.0 M
136 HNO_3. The resulting solution enriched in lanthanoids was removed from the Shielded Cells
137 Facility and analyzed using gamma-ray spectroscopy to verify removal of high-activity fission
138 products.

139 Aliquots of each 50 mL solution (1 mL each, except for sample A where only 0.1 mL
140 was taken due to the higher total activity of the lanthanoids) were shipped to UMD for separation
141 of the individual lanthanoids. This separation, which was carried out in a vented laminar flow
142 hood, certified for radioactive materials, involved the use of AG-50W x8 200-400 mesh resin in
143 the NH_4^+ form loaded into a 0.3 cm x 28 cm quartz glass column and alpha-hydroxyisobutyric
144 acid (α-HIBA) in three strengths 0.15 M, 0.225 M, and 0.53 M all buffered to pH 4.7 using
145 NH_4OH (10, 11, 12).

146 The aliquots were dried and reconstituted in 0.2 mL of 0.15 M α-HIBA and loaded onto
147 the columns. Following an 8 mL wash of 0.15 M α-HIBA, the Gd cut was collected in 4.25 mL
148 of 0.15 M α-HIBA. A subsequent 8 mL wash of 0.15 M α-HIBA removed most of the Eu, with
149 the lighter lanthanoids later collected in 10 mL of 0.53 M α-HIBA. The column was then
150 sequentially treated with 6 M HCl, 18 MΩ H_2O, 7 M NH_4OH, and equilibrated with 0.225 M α-
151 HIBA. The 10 mL aliquot containing the lighter lanthanoids was dried down and the α-HIBA
152 digested using aqua regia. The remaining material was reconstituted in 0.2 mL 0.225 M of α-
153 HIBA and loaded onto the re-equilibrated column. The column was washed with 3 mL of 0.225

M of α-HIBA. Sm was recovered with 3.75 mL of 0.225 M of α-HIBA. The column was washed with 9 mL of 0.225 M of α-HIBA. Finally, Nd was collected with 4.5 mL of 0.225 M of α-HIBA. The Gd, Sm, and Nd aliquots were dried down and the α-HIBA digested using aqua regia. These final solutions were then dried down and reconstituted in 2 mL 0.8 M HNO_3 for Sm and Nd, while Gd was reconstituted in 1 mL 0.8 M HNO_3. Aliquots of the resulting solutions of Nd and Sm were taken for MC-ICP-MS analyses while the entire 1 mL Gd solution was analyzed.

Reference materials for this work consisted of BHVO-2 (a basalt powder reference material from the USGS), a 600 ppb (g/g) solution of lanthanoids (Ce through Dy) in 2 M HCl, created via dilutions of concentrated elemental stock solutions from Alfa Aesar (referred to as UMD1), as well as a total procedure blank. Two 0.05 g samples of BHVO-2 were digested using a mixture of HNO_3 and HF with 100 μL of $HClO_4$ in a sealed 15 mL PTFE beaker at 180°C. The solution was then dried down and the samples were reconstituted in 6 M HCl, sealed, and heated for 24 hours. This process was repeated twice and the final dry material was dissolved in 2 mL of 2 M HCl. The reference samples were processed through all chromatographic separations identically to the fuel rod solutions. In addition a blank (UMD Blank) was prepared and treated identically to the reference samples. All separations for the reference materials and UMD Blank were performed in the clean laboratory at UMD and a schematic of the separation process is shown in Figure 1.

MC-ICP-MS

Isotopic analyses of Nd and Sm were conducted at the Department of Geology, UMD using a Nu Plasma MC-ICP-MS with operating parameters reported in Table 2. The instrument

176 was coupled to an Aridus I desolvating nebulizer with an uptake rate of 50 μL/min. The sample
177 measurement conditions for Nd consisted of 5 blocks of 20 5-second integrations preceded and
178 followed by a 15-second integration of the background at half a mass unit above the peak. Those
179 for Sm included 4 blocks of 20 7-second integrations (1 second for Cycle 2) and 15-second
180 integration for backgrounds. Gd is not reported due to the measured signal being below the
181 quantification threshold for the fuel samples. Reasons for this will be discussed later.

182 Instrument induced mass fractionation was corrected with the standard-sample-standard
183 bracketing method using the UMD1 solution as the standard and the BHVO-2 samples as a
184 quality control check to determine the accuracy of the correction method. In addition the AMES
185 Nd isotopic standard was diluted to give similar signal intensity as the samples and which was
186 analyzed to provide an additional QC control for Nd. For the UMD1 solutions it was assumed
187 $^{146}Nd/^{144}Nd = 0.7219$ and $^{148}Sm/^{154}Sm = 0.49419$ as the internal correction values (13). Total
188 intensity of the ion beams for samples and UMD1 deviated by no more than 15 % during the
189 analysis with total Nd and Sm intensities of ~6.4 V and 2.1 V, respectively, with all faraday cups
190 having 1 x 10^{11} Ω resistors.

191 A number of short-lived isobaric interferences for Nd and Sm exist in addition to
192 naturally occurring isobars. The interferences for Nd include ^{142}Ce and $^{144,148,150}Sm$, while Sm
193 has interferences from $^{144,148,150}Nd$ and $^{151,152,154}Eu$. Isobaric interferences for Nd were
194 monitored during Nd analysis by using intensities of ^{140}Ce and ^{147}Sm as they do not have any
195 isobaric interferences. The intensities of ^{140}Ce and ^{147}Sm and averaged approximately 3 mV and
196 0.3 mV, respectively, while Sm interferences were monitored using ^{145}Nd and ^{153}Eu and were
197 approximately 0.01 mV and 0.1 V respectively.

198

Results

Chemistry yields for Nd, Sm, and Gd were calculated by comparing the total ion intensities of the UMD1 samples to stock solutions of known concentration prepared gravimetrically prior to isotopic analysis. This resulted in yields of 59 ± 4%, 53 ± 10%, and 43 ± 6% (2σ) for Nd, Sm, and Gd respectively. Yields from BHVO-2 samples fall within these ranges with the noticeable exception of a calculated 80% yield for Gd. Concentrations for Nd and Sm in the MTR fuel solutions and their respective chemistry blank solutions contained in the Shielded Cells Facility at SRNL are presented in Table 3. Analysis of Gd in the MTR solutions showed no detectable signal.

The isotopic abundances of Nd and Sm in the MTR fuel and BHVO-2 were corrected using the mass fractionation correction terms derived from the UMD1 standards and the results are given in Table 4. The overall external reproducibility of the MC-ICP-MS is approximately 0.02% (2σ) and the bias introduced via the bracketing correction method is 0.01-0.4 %. Isobaric interferences from Nd on Sm were indistinguishable from measurement uncertainty. However, Sm interferences on Nd isotopic ratios required corrections on ^{148}Nd and ^{150}Nd. To perform this correction, the ^{148}Sm/^{147}Sm and ^{150}Sm/^{147}Sm ratios from the respective fuel assemblies were used in conjunction with the measured ^{147}Sm monitored during Nd measurement to correct for ^{148}Sm and ^{150}Sm on ^{148}Nd and ^{150}Nd for all of the fuel assemblies. The final abundances for Nd and Sm isotopes with the overall reproducibility, the bracketing bias, and isobaric interferences incorporated into the uncertainties are presented in Table 5.

Discussion

Yields and Blanks

Chemistry blank values for the fuel rod samples are far higher than those of the chemistry blank from UMD as expected from the hot cell work. The fuel rod blanks range from 2 μg to 1 ng while the UMD blanks were between 2-3 pg. This difference in magnitude confirms that the separation process at UMD did not introduce a notable amount of natural lanthanoids which could have affected the isotopic signature of the fuel assemblies. Isotopic measurements of Blank A and Blank C show isotopic composition identical to their corresponding fuel samples, making it unlikely that the blanks were contaminated by legacy Savannah River Site processes that had been previously sampled in the Shielded Cells facility. The isotopic composition of Blank B was not measureable due to insufficient signal intensity for faraday cup measurements. Ion counters were not logistically practical due to concerns over residual radioactive contamination after the analyses and were not used for radioactive sample measurements. The UMD Blank, however, was measured using the ion counters and they were calculated assuming a natural isotopic composition. The similarity between the fuel rod isotopic composition and their respective blanks discredits the possibility of contamination from natural sources and instead implies that the blanks have been cross contaminated with the fuel rod material. Blank A has the highest amount of Nd and Sm while Blank B and C are approximately 10-1000 times lower. It is unclear where and how the contamination events occurred, but the contamination had no notable effect on the final reported isotopic compositions of the fuel assemblies.

Based on fission yields of Gd compared to Nd and Sm, Gd should have been produced in the sample however, no detectable signal was observed for Gd. Neutron induced fission of ^{235}U produces around 0.05% Gd and 3% Sm (3). The concentration of Sm in the MTR samples at UMD was on the order of micrograms. Therefore, Gd should have been present in the sample at

244 the ng level, well above the pg limit of detection when taking into account the 43% recovery.
245 Therefore, the most probable reason for the lack of a Gd signal is a loss of the Gd cut from the
246 2^{nd} stage column. Unfortunately, operational restrictions on re-sampling the 50 mL solutions at
247 SRNL resulted in an inability to reevaluate the procedure and determine the isotopic composition
248 of Gd.

249 Another possible explanation for this low yield is that the Gd peak was eluted before or
250 after the collection step. The Gd cut elutes over 4 mL of 0.15 M α-HIBA so the possibility that
251 the entire Gd peak was not collected is unlikely, especially when compared with the successful
252 collection of Gd from the BHVO-2 samples.

253

254 *Nd and Sm Isotope Abundances*

255 To determine if the burn-up of ^{235}U has any effect on the isotopic composition of the fuel
256 assemblies, the ratio of fuel assemblies A:B and C:B are presented in Figures 2 and 3 with no
257 decay corrections. In the case of Nd (Fig. 2) the final ^{235}U enrichment seems to have no effect
258 on the production of Nd isotopes, with the exception of ^{142}Nd, ^{143}Nd, and ^{144}Nd. All of the listed
259 exceptions could be explained due to differences in neutron fluence during reactor operation as
260 reactions $^{141}Pr(n,\beta)^{142}Nd$ and $^{143}Nd(n,\gamma)^{144}Nd$ have thermal neutron capture cross sections of 11
261 and 325 barns, respectively (14). While the ^{141}Pr capture cross section is small, the fission
262 production of ^{141}Pr is relatively high at ~5.9% (3), which results in a large amount of available
263 ^{141}Pr for neutron capture reactions to produce ^{142}Nd. The thermal capture cross section for
264 $^{142}Nd(n,\gamma)^{143}Nd$ is similar to $^{141}Pr(n,\beta)^{142}Nd$ at 18 barns (14), however, due to the much lower
265 total amount of available ^{142}Nd produced through neutron capture of ^{141}Pr and from fission (10^{-9}

%) results in a negligible amount of ^{142}Nd being converted to ^{143}Nd. Therefore, higher neutron fluence has the overall effect of increasing the abundance of ^{142,144}Nd in the nuclear fuel while decreasing the abundance of ^{143}Nd.

The other isotopes of Nd (mass 145 through 150) do not show any obvious differences between the fuel assemblies. This is expected due to the low thermal neutron capture cross sections (50, 1.5, 2.6, and 1.0 barns for ^{145}Nd, ^{146}Nd, ^{148}Nd, and ^{150}Nd, respectively (14)) for these isotopes. Additionally, based on the reported burnup history of these fuels, it appears that the initial and final ^{235}U enrichments of the fuel have no notable influence on ^{145,146,148,150}Nd abundances.

The graph of fuel rod A:B and C:B for Sm (Figure 3) notably shows more isotopic variation between the fuel samples for Sm as compared to Nd. The cause of the fluctuations for ^{147}Sm, ^{148}Sm, ^{149}Sm, ^{150}Sm, and ^{151}Sm is likely due to thermal neutron captures on ^{147}Sm, ^{149}Sm, and ^{151}Sm which have thermal capture cross sections of 57, 40,000, and 15,000 barns, respectively (14). Further evidence for neutron capture reactions causing the Sm isotopic differences between the fuel rods is seen in the enrichments in A:B mirrored by depletions in C:B and vice versa.

Interestingly, ^{144}Sm is unique in that both fuel assemblies A and C are depleted relative to fuel rod B. As this isotope is not produced directly via ^{235}U fission or neutron capture reactions, the ^{144}Sm present is a remnant of natural Sm in either the nuclear fuel or the cladding material mixing with fission produced Sm which contains no ^{144}Sm. To determine if the ^{144}Sm is actually a sign of contamination from natural sources on the analyzed aliquots a simple calculation was used to determine the amount of natural Sm necessary to create the same isotopic composition in the fuel assemblies assuming an initial starting point of no Sm present. The resulting blank

289 contribution would need to be between 5 and 1500 times greater in the fuel samples than what
290 was measured in the respective fuel blank samples. With the assumption of the ^{144}Sm originating
291 solely from natural Sm, there is approximately 4(1), 5(2), and 0.7(2) μg natural Sm in fuel
292 assemblies A, B, and C, respectively. These masses result in Sm concentrations of 3(1), 3(2),
293 and 0.6(2) ppm in the aluminum cladding which are less than the 5 ppm the American Society
294 for Testing and Materials (ASTM) sets for high purity Al cladding (15). If the natural Sm came
295 from the nuclear fuel instead of the cladding the concentrations would range between 11(3),
296 35(11), and 2.6(9) ppm, well above the <3 ppm total lanthanoids the ASTM sets for enriched
297 uranium (16). While assembly C is close to the limit, the 2.6(9) ppm value is for Sm only and it
298 is reasonable to assume that the other lanthanoids would be present in similar quantities which
299 would make the assembly not meet ASTM standards, therefore unlikely that the natural material
300 came from the fuel itself.

301

302 **5.5 Conclusions**

303 A two stage separation technique involving cation exchange chromatography has successfully
304 separated Nd and Sm from three MTR spent nuclear fuels. The isotopic abundances of Nd and
305 Sm are distinctively non-natural and show depletions and enrichments in isotope pairs that have
306 large cross sections for thermal neutron absorption and corresponding product nuclei with low
307 thermal neutron absorption cross section. Signs of residual natural ^{144}Sm originating from the
308 cladding of the fuel samples are observed when comparing the Sm isotopic composition between
309 the three fuel assemblies. The isotopic composition of Nd, with the exception of ^{142}Nd, ^{143}Nd,
310 and ^{144}Nd, is indistinguishable between the three nuclear fuel assemblies and is identical to
311 predicted abundances derived from ^{235}U fission. Sm isotopic compositions are more varied

between the fuels, which is attributed to differences in reactor neutron fluences and natural Sm contamination in the fuel cladding. Overall, the ability to isolate and analyze Nd and Sm from spent nuclear fuel using readily available materials and identified differences between three MTR spent nuclear fuels has been demonstrated.

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Table 1: Uranium enrichments of the three spent MTR nuclear fuels.

	Initial ^{235}U	Final ^{235}U
A	93%	70%
B	93%	75%
C	45%	33%

Table 2: Instrument operational conditions used for MC-ICP-MS analysis of MTR samples.

Nu Plasma HR MC-ICP-MS parameters	
RF Power	1300 W
Reflected Power	5 W
Accelerating Voltage	4000 V
Cool Gas Flow	13 L min^{-1} Ar
Auxiliary Gas Flow	1 L min^{-1} Ar
Sweep Gas Flow[a]	2.75 L min^{-1} Ar
N_2 Gas	10 mL min^{-1} N_2
Aspiration Rate	50 μL min^{-1}

Collector[b]	H5	H4	H3	H2	H1	Ax	L1	L2	IC0	L3
Nd	150	148	147	146	145	144	143	142		140
Sm[c]	154|5	152|3	151|2	150|1	149|50	148|9	147|8	146|7		144|5

[a]Gas flows are set with an Aridus I

[b]H/Lx cups refer to Faraday cups, IC0 is an ion counter

[c]Sm analyses involved two cycles: Cycle 1 | Cycle 2

Table 3: Concentration of Nd and Sm (μg/mL) in stock solutions at SRNL and total chemistry blank obtained at UMD. Blanks with no uncertainties correspond to blanks below limit of detection.

	Nd	Sm
Fuel A	$1.3(3) \times 10^{2}$	14(4)
Blank A	$3.8(9) \times 10^{-2}$	$1.3(4) \times 10^{-2}$
Fuel B	$6(1) \times 10^{2}$	13(4)
Blank B	1×10^{-4}	2×10^{-5}
Fuel C	$3.1(7) \times 10^{2}$	6(2)
Blank C	$1.2(3) \times 10^{-2}$	4×10^{-5}
UMD Blank	2 pg	3 pg

Table 4: Nd and Sm isotopic ratios in MTR samples and natural materials with 2σ uncertainties.

Sample (Nd)	$^{142}Nd/^{144}Nd$	$^{143}Nd/^{144}Nd$	$^{145}Nd/^{144}Nd$	$^{146}Nd/^{144}Nd$	$^{148}Nd/^{144}Nd$	$^{150}Nd/^{144}Nd$	Nd Beam (V)	^{147}Sm (V)	^{140}Ce (V)
A	0.00955(1)	0.61910(5)	0.50918(4)	0.46731(6)	0.24410(6)	0.08862(3)	5.39	0.00303	0.00026
B	0.00724(1)	0.70851(9)	0.55239(2)	0.47651(7)	0.25761(7)	0.09564(3)	4.80	0.00023	0.00036
C	0.00215(1)	0.93197(9)	0.63756(4)	0.52532(7)	0.29817(8)	0.10887(4)	5.42	0.00147	0.00008
BHVO-2 1	1.14038(6)	0.51264(2)	0.34861(1)	0.72278(4)	0.24213(3)	0.23718(3)	6.29	0.00258	0.01340
BHVO-2 2	1.14193(5)	0.51301(2)	0.34839(1)	0.72180(3)	0.24147(2)	0.23621(2)	7.18	0.00003	0.00142
Ames (n=2)	1.14173(5)	0.51213(2)	0.34840(1)	0.72190[a]	0.24154(1)	0.23630(2)	6.69	0.00000	0.00009
UMD1 (n=3)	1.14173(40)	0.51239(11)	0.34839(5)	0.72190[a]	0.24156(3)	0.23630(6)	7.03	0.00002	0.00125

Sample (Sm)	$^{151}Sm/^{154}Sm$	$^{144}Sm/^{154}Sm$	$^{147}Sm/^{154}Sm$	$^{148}Sm/^{154}Sm$	$^{149}Sm/^{154}Sm$	$^{150S}m/^{154}Sm$	$^{152}Sm/^{154}Sm$	Sm Beam (V)	^{146}Nd (V)	^{153}Eu (V)
A	0.1676(2)	0.00206(10)	11.7550(5)	3.5148(7)	0.1732(2)	8.9301(36)	4.5568(6)	2.21	0.00007	0.7
B	0.3687(3)	0.00475(14)	21.185(16)	3.4269(11)	0.4646(2)	12.7430(69)	7.8544(13)	2.28	0.00010	0.1
C	0.6589(5)	0.00134(14)	22.336(14)	1.5066(3)	0.6811(2)	11.8466(44)	7.9488(7)	2.19	0.00002	0.2
BHVO-2 1	n/a[c]	0.13537(4)	0.6599(1)	0.49466(4)	0.60811(8)	0.32472(4)	1.17586(3)	2.16	0.0008	0.02
BHVO-2 2	n/a[c]	0.13538(4)	0.65984(9)	0.49452(3)	0.60799(6)	0.32465(4)	1.17574(3)	2.18	0.0001	0.03
UMD1 (n=4)	n/a[c]	0.13527(4)	0.65937(6)	0.49419[b]	0.60769(3)	0.32450(6)	1.17549(3)	2.01	0.00001	0.0003

[a] $^{146}Nd/^{144}Nd = 0.7219$ for mass fractionation correction

[b] $^{148}Sm/^{154}Sm = 0.49419$ for mass fractionation correction

[c] No ^{151}Sm exists in natural samples

Table 5: Isotope abundances of Nd and Sm in the three MTR fuels.

Nd Isotope	A	B	C
^{142}Nd	0.3250(3)%	0.2336(2)%	0.0615(2)%
^{143}Nd	21.073(5)%	22.871(6)%	26.597(6)%
^{144}Nd	34.038(1)%	32.280(1)%	28.539(1)%
^{145}Nd	17.332(2)%	17.831(2)%	18.195(2)%
^{146}Nd	15.907(2)%	15.382(2)%	14.992(2)%
^{148}Nd	8.309(2)%	8.316(3)%	8.509(3)%
^{150}Nd	3.016(1)%	3.087(1)%	3.107(1)%
Sm Isotope	MTR 2009	MTR 2010	MTR 2011
^{147}Sm	39.06(2)%	45.03(4)%	48.58(6)%
^{148}Sm	11.678(4)%	7.284(3)%	3.277(1)%
^{149}Sm	0.576(1)%	0.988(1)%	1.481(1)%
^{150}Sm	29.67(1)%	27.09(2)%	25.77(1)%
^{151}Sm	0.557(1)%	0.784(1)%	1.433(1)%
^{152}Sm	15.140(4)%	16.695(7)%	17.288(6)%
^{154}Sm	3.3225(3)%	2.126(1)%	2.175(1)%

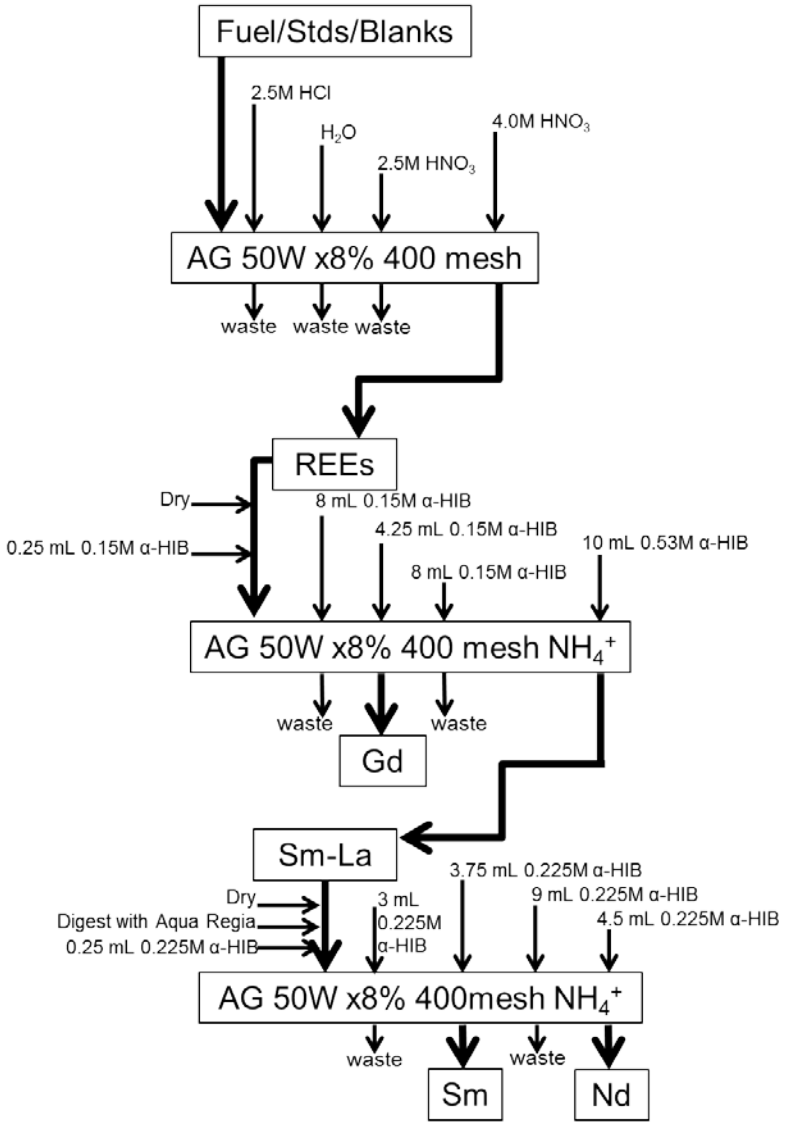

Figure 1: Separation schematic for isolating Gd, Sm, and Nd. Bold lines denote analyte path.

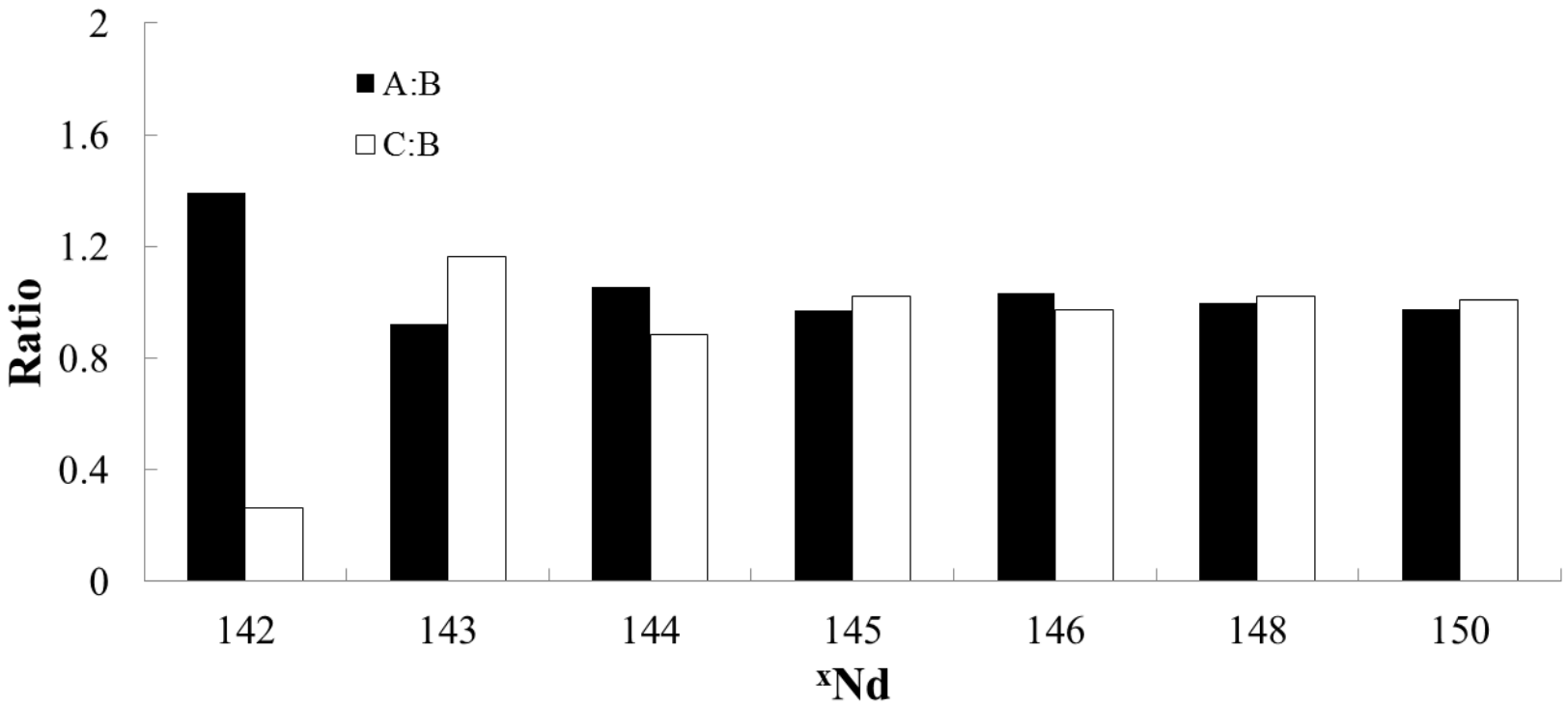

Figure 2: Ratio of Nd isotope abundance of fuel rods A and C to B. Uncertainty is indistinguishable at the above scale.

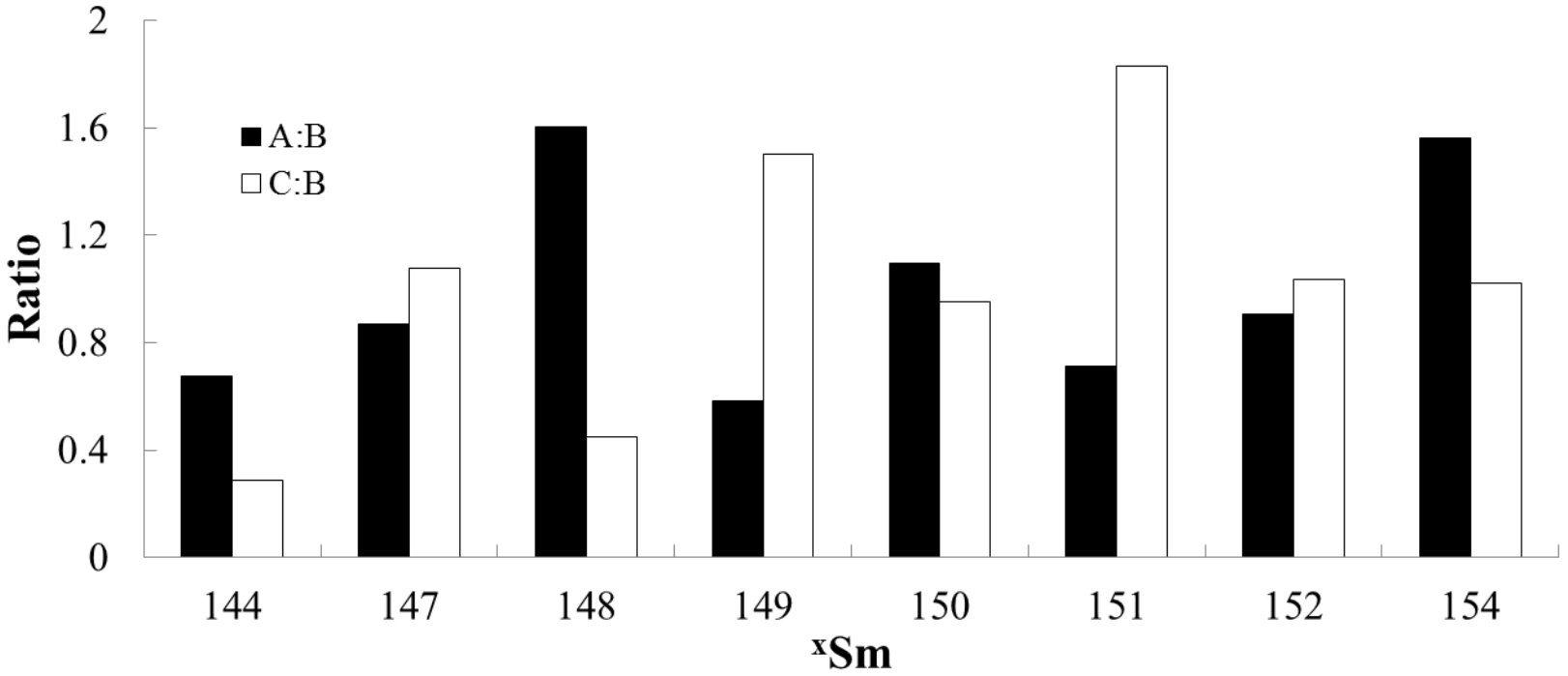

Figure 3: Ratio of Sm isotope abundance of fuel rods A and C to B. Uncertainty is indistinguishable at the above scale.