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FY13 Summary Report on the Augmentation of the Spent Fuel Composition Dataset for Nuclear Forensics: SFCOMPO/NF

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March 2014

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Pacific Northwest National Laboratory
Richland, Washington 99352

Abstract

This report documents the FY13 efforts to enhance a dataset of spent nuclear fuel isotopic composition data for use in developing intrinsic signatures for nuclear forensics. A review and collection of data from the open literature was performed in FY10. In FY11, the Spent Fuel COMPOsition (SFCOMPO) Excel-based dataset for nuclear forensics (NF), SFCOMPO/NF was established and measured data for graphite production reactors (PRs), Boiling Water Reactors (BWRs) and Pressurized Water Reactors (PWRs) were added to the dataset and expanded to include a consistent set of data simulated by calculations. A test was performed to determine whether the SFCOMPO/NF dataset will be useful for the analysis and identification of reactor types from isotopic ratios observed in interdicted samples.

The FY12 program resulted in the addition of measured data for CANDU reactors, MAGNOX reactors, VVERs, and RBMKs to the dataset. This measured data was also expanded by some calculation models to include a consistent set of data. Finally, comparisons were made with the expanded SFCOMPO/NF dataset to see if it will be useful for the analysis and identification of reactor types from isotopic ratios observed in interdicted samples.

The FY13 efforts expanded upon the FY12 outcomes by updating the existing SFCOMPO/NF dataset to incorporate the reported uncertainty information associated with the measured data, and adding the sources of the measured data directly to the dataset. The uncertainty data was used to compare the spread in data and the uncertainties. Additionally, preliminary correlations between fuel burnup, initial enrichment, and isotopic ratios of interest were examined.

The augmented SFCOMPO/NF dataset now includes measurements, their uncertainties where available, and some model data for PWRs, BWRs, graphite PRs, CANDU reactors, MAGNOX reactors, VVERs and RBMKs to facilitate testing the database for use in discriminating the reactor of origin. Comparisons are made using various ratios of the plutonium isotopes ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu. These comparisons demonstrate that the dataset can be used to differentiate materials originating in graphite PRs, CANDU reactors, MAGNOX reactors, VVERs and RBMKs compared to material that originate in light water reactors (LWRs). The results also show trends that will be able to support the distinction between BWRs and PWRs (two types of LWRs). There may be additional comparisons that could be made in the future using other isotopes such as minor actinides or trace fission products that will allow for further identification within a reactor type to identify fuel enrichment, burnup, age and perhaps country of origin.

Discussions are also presented to identify and prioritize any additional measured and calculated data that need to be added to the SFCOMPO/NF dataset to broaden the number and type of reactor systems that are included. There is a need for some additional modeling data to fill in gaps in the measured data and enable the discrimination of fuel at a finer level of detail. Most of the measured data that is believed to be available in the open literature has been identified and integrated into SFCOMPO/NF. Measured data are very limited for some reactor types. There is still a need for new measured data to validate the reactor models used for augmenting SFCOMPO/NF. Code validation is necessary to determine the degree of confidence (uncertainty) that the calculated values accurately reflect reality. During FY12, Neutron Activation Analysis (NAA) was performed on two samples dissolved from the high burnup commercial UO_2 fuel Approved Testing Material (ATM-109) for identification of the stable and quasi-stable noble

metal nuclides. Mass quantities of ^{98}Mo, ^{100}Mo, ^{102}Ru, ^{104}Ru and ^{103}Ru were obtained with an uncertainty level of two-sigma. Sources of uncertainty include counts, efficiency, gamma ray intensity, decay constant, cross section, neutron flux, irradiation time and decay time.

Acronyms and Abbreviations

ADSNF	Assay Data for Spent Nuclear Fuel
AGR	Advanced Gas-cooled Reactors
ARIANE	Actinides Research in a Nuclear Element
ATM	Approved Testing Material
BUC	Burnup Credit Criticality Safety
BWR	Boiling Water Reactor
CANDU	Canada Deuterium Uranium Reactor
E/C	Ratio of experimental value to calculated value
EG	Expert Group
FBR	Fast Breeder Reactor
GCHWR	Gas Cooled Heaver Water Reactor
GWd/MTU	Gigawatt Days/Metric Tonne Uranium
HTGR	High Temperature Gas-Cooled Reactor
ICPMS	Inductively Coupled Plasma-Mass Spectrometry
IDMS	Isotope Dilution Mass Spectrometry
JAEA	Japan Atomic Energy Agency
JAERI	Japan Atomic Energy Research Institute
KKG	Kernkraftwerk Gosgen
LWCHWR	Light Water Cooled Heavy Water Reactor
LWGR	Light Water Graphite Reactor
LWR	Light Water Reactor
MAGNOX	Magnesium Non-Oxidizing Alloy
NAA	Neutron Activation Analysis
NEA	Nuclear Energy Agency
NETL	Nuclear Engineering Teaching Laboratory
NF	Nuclear Forensics
NTNFC	National Technical Nuclear Forensics Center
OECD	Organization for Economic Cooperation and Development
ORIGEN	Oak Ridge Isotope GENeration
PHWR	Pressurized Heavy Water Reactor
PNNL	Pacific Northwest National Laboratory
PR	Production Reactor
PWR	Pressurized Water Reactor
RBMK	Reaktory Bolshoy Moschnosti Kanalniy, or “High power channel-type reactor” (A Russian Light Water Graphite Reactor)
SCALE	Standardized Computer Analysis for Licensing Evaluation

SFCOMPO	Spent Fuel COMPOsition database
SGHWR	Steam Generating Heavy Water Reactor
SNF	Spent Nuclear Fuel
SOAR	State Of the Art Report
VVER	Vodo-Vodyanoi Energetichesky Reaktor, or “Water-water energetic reactor” (Pressurized Water Reactor of Russian design)
WPNCS	Working Party on Nuclear Criticality Safety

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1.0 Introduction

There are limited databases of spent fuel radionuclide inventories suitable for comparison against interdicted materials or for validating and verifying computational models. Leveraging its participation on international committees, Pacific Northwest National Laboratory (PNNL) significantly increased the size and quality of experimental datasets available for nuclear forensics purposes.

PNNL had some successes in identifying measured inventory data for a number of reactor types as documented in the FY10 final report. The previous FY10 summary report also discussed the importance of integrating model data into the dataset to provide a complete set of reactor data for analysis.

The FY11 summary report documented the expansion of the existing inventory data in the SFCOMPO/NF (SFCOMPO/Nuclear Forensics) dataset in order to develop approaches to identifying reactor of origin. Additionally, PNNL migrated the existing SFCOMPO data and established the special purpose dataset, SFCOMPO/NF independent of the source database. The content and layout of SFCOMPO/NF is further described in Section 4. Specific measurements and model calculations that were incorporated into SFCOMPO/NF in FY11 and FY12 are described in Section 5 and Section 6.

The FY12 effort involved expanding this SFCOMPO/NF dataset with data from additional reactor types in order to enhance its value to nuclear forensics. Appendix A summarizes reactor samples that were considered to be added which were not in the SFCOMPO dataset. The FY12 work is a significant step to expand the data by including measured and simulated isotopic inventories of spent fuel from a variety of reactor types including: Canada Deuterium Uranium (CANDU), MAGNOX, **V**odo-**V**odyanoi **E**nergetichesky **R**eaktor (VVER) and **R**eaktor **B**olshoy **M**oshchnosti **K**analniy, "High Power Channel-type Reactor" (RBMK).

The FY13 work expanded upon the FY12 outcomes by updating the existing SFCOMPO/NF dataset to incorporate the reported uncertainty information associated with the measured data, while adding the sources of the measured data directly to the dataset. The uncertainty data was used to compare the spread in data and the uncertainties. Additionally, correlations between fuel burnup, initial enrichment, and isotopic ratios of interest were examined and a simple second order least squares fit was applied to the plutonium ratio measurement data of each reactor type in an attempt to characterize that reactor type through those ratios.

The intended use of these data is to investigate the use of ratios of key plutonium isotopes to discriminate materials derived from these different reactor types. Known attributes of these plutonium ratios for other reactors, such as graphite PRs, CANDU reactors, MAGNOX reactors, VVERs and RBMKs provided expectation that the distinction for these and the group of PWR and BWR fuels would be clear. The objective was to verify this expectation and then to look for additional trends that would further discriminate PWRs from BWRs. Future work will also expand to include discriminating signatures from within a reactor type to identify specific operating conditions that may yield information regarding country of origin, age, etc.

The principle data modification to SFCOMPO/NF was to include the integration of measured and simulated isotopic data for graphite-moderated light-water cooled plutonium PRs, CANDU reactors, MAGNOX reactors, VVERs and RBMKs. The addition of these data permitted the use of the

SFCOMPO/NF dataset to test the assumption that spent fuel isotopic inventory data can be compared against measurements of interdicted material to identify the reactor type of origin.

2.0 Background

Databases of experimentally derived spent fuel parameters have been historically developed to facilitate the verification and validation of computational models. In the case of nuclear forensics, these databases might provide the essential information to compare against interdicted nuclear materials measurements and allow the origin of the material to be determined.

In 1993, the Japan Atomic Energy Research Institute (JAERI, the organization is now known as the Japan Atomic Energy Agency, JAEA) began development of a database for the isotopic composition of commercial spent nuclear fuel (SNF). Data for this database, known as SFCOMPO (Spent Fuel Compositions) were collected from open literature.[1] In 2002, ownership and responsibility for maintaining and updating SFCOMPO was transferred to the OECD/NEA in order to provide for a more international framework for its development.[2] SFCOMPO currently contains measured isotopic compositions from 14 reactors (7 Pressurized Water Reactors (PWRs) and 7 Boiling Water Reactors (BWRs)) in four countries.

SFCOMPO was originally developed to support the validation of computer codes and models used to calculate the isotopic composition of light water reactor fuels for use in evaluating criticality safety for spent fuel operations. The measured data in SFCOMPO as of FY10 were a compilation of information available in the open literature for PWR and BWR fuels.

While SFCOMPO has been used for validating such codes as ORIGEN (Oak Ridge Isotope GENeration) and SCALE (Standardized Computer Analysis for Licensing Evaluation), there are significant limitations to the current database that affect its usefulness for the purpose of nuclear forensics. A database of spent fuel isotopic data would need to include data for a broad variety of reactor types to have a significant impact in evaluating interdicted material. It is also necessary to have enough data to cover the range of operating parameters typical of each reactor type. Significant parameters include burnup, fuel enrichment/composition, and fuel age or cooling time. Other useful parameters include the history of the reactor power, and the poisons used (See Table 1).

PNNL began an effort in FY10 to identify additional measured data that could be incorporated into SFCOMPO to expand the existing range of operating parameters for PWR and BWRs as well as to expand the database to include additional types of reactors. There are a wide variety of power reactors in operation today as identified in Appendix B. The principle reactor types of interest include VVERs, RBMKs, CANDU reactors and MAGNOX reactors. PNNL identified a fair amount of data that is unclassified and available in open literature which could be incorporated into SFCOMPO[3] to expand its value for use in nuclear forensics.

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[1] Kurosawa, M, Y Naito, H Sakamoto, and T Kaneko. 1997. *The Isotopic Compositions Database System on Spent Fuels in Light Water Reactors (SFCOMPO)*. Report No. JAERI-Data/Code 96-036, Japan Atomic Energy Research Institute.

[2] SFCOMPO is an international database of spent fuel isotopic data that is maintained by the Nuclear Energy Agency of the Organization for Economic Cooperation and Development (OECD/NEA) http://www.oecd-nea.org/sfcompo/

[3] Hanson BD, CZ Soderquist, AJ Casella, AM Casella, and AM Johnsen. 2012. *Analysis of High-Burnup and MOX Fuel for Inclusion in International Spent Fuel Databases* . PNNL-21254, Pacific Northwest National Laboratory, Richland, WA.

Additionally, concerns have been expressed that the data included in SFCOMPO have not been subjected to a detailed peer review. There is an international effort lead by the OECD (Organization for Economic Cooperation and Development)/NEA (Nuclear Energy Agency) Expert Group (EG) on Assay Data of Spent Nuclear Fuel (ADSNF)[4] within the Working Party on Nuclear Criticality Safety (WPNCS) to develop and implement a peer review/evaluation process for data included in SFCOMPO. As part of this evaluation process, the EGADSNF will also develop recommended uncertainty values. While we agree that proper peer review is essential for a useable database, we believe the NTNFC (National Technical Nuclear Forensics Center) program is best served by monitoring the international effort rather than incurring the expense of developing an independent process.

The development of a more comprehensive spent fuel isotopic database included a component to address the measurement of new data and the concern that there are no set standards for how the isotopic analyses are to be performed and for reporting the uncertainties. The FY11 report[5] provided the results for the chemical dissolution and analysis of a spent fuel samples. This effort was reduced for FY12[6] and some preliminary results were presented related to assessing the usefulness of neutron activation analysis as a potential measurement tool for use with dissolved fuel samples.

[4] http://www.oecd-nea.org/science/wpncs/ADSNF/

[5] Brady Raap MC, BA Collins and CJ Francy. 2012. *FY11 Summary Report on the Augmentation of the Spent Fuel Composition Database for Nuclear Forensics: SFCOMPO/NF*. PNNL-21939, Pacific Northwest National Laboratory, Richland, WA.

[6] Brady Raap MC, BA Collins, CJ Francy and Ann Doherty. 2013. *FY12 Summary Report on the Augmentation of the Spent Fuel Composition Database for Nuclear Forensics: SFCOMPO/NF*. PNNL-22437, Pacific Northwest National Laboratory, Richland, WA.

3.0 Isotopic Compositions

The ultimate objective is to develop statistical models using both experimental and calculated data. These models will be used for the identification of key reactor characteristics that could be a credible source of specific nuclear materials obtained from the investigation of interdicted or post detonation evidence. The usefulness and reliability of these models will be dependent on the quality and quantity of data upon which they are based. Another factor necessary for the successful use of these models is that they represent the range of expected normal and known operating conditions for the specific reactor type. The quantities of measured spent fuel isotopic data which are available in the open literature are limited with respect to the range of initial fuel compositions, assembly and reactor design, and other operating conditions. In order to protect proprietary information, it is common to report data in the form of ratios most often cited as E/C values (ratio of experiment to calculated values). This type of "measured" data can also be used to greatly expand the range of reactor types and operating parameters that could be addressed in this statistical modeling. It was recommended in the FY10 report[7] that calculational models (i.e. computer codes) be used to produce simulated data to represent the full range of applicability for each reactor type.

The current effort for the development of SFCOMPO/NF has been to add measured data uncertainties and explicit reference link to the data file.

3.1 Experimental Data

The Spent Fuel Composition (SFCOMPO) database maintained by the OECD/NEA is the largest compilation of open data for Pressurized Water Reactor (PWR) and Boiling Water Reactor (BWR) fuels. This database and a subset of the experimental data identified in the FY10 report[7] were the source of data used to develop SFCOMPO/NF. Additional measurements for graphite PRs[8] were identified for incorporation into SFCOMPO/NF. In FY12, some additional data for CANDU reactors, MAGNOX reactors, VVERs and RBMKs was added to SFCOMPO/NF.

3.2 Calculated Data

The analyses of fuel burnup and depletion, as have traditionally been performed for reactor and fuel design and performance evaluations, have been focused on a select and limited set of nuclides- primarily the major fissile actinides. More recently, the evolution of analysis techniques for away-from-reactor fuel safety calculations has identified the need and capability to predict the composition of irradiated fuel for a larger set of isotopes. Concurrent with the evolution of analysis capabilities, the need for additional experimental data to validate these isotope inventory calculations was realized. The quantity of non-proprietary data to fill this need has been limited. Various groups have made attempts to compile and compare the available open data and analysis techniques. The OECD/NEA Expert Group (EG) on Burnup

[7] Hanson, BD, AM Johnsen, BA Collins, PP Schonewill, MC Brady Raap, and GL Swearingen. 2012. *FY10 Summary Report Augmentation of International Spent Fuel Databases.* PNNL-21469, Pacific Northwest National Laboratory, Richland, WA.

[8] Toffer H & Kupinski AF, *"Experimental Isotopic Analysis of Point Exposure Data in Hanford Production Reactor Fuels"*. DUN-7243, Douglas United Nuclear, Inc. Richland, WA, September 8, 1970

Credit Criticality Safety (BUC)[9] and the Expert Group (EG) on Assay Data for Spent Nuclear Fuel (ADSNF) are the predominant groups working with BWR and PWR data.

The development of models to predict the isotopic compositions in used fuel requires specific knowledge of the (1) initial fuel composition and (2) operating (exposure) history of the fuel (see Table 1).

Table 1. Examples of Data Used in Isotopic Predictions

Initial Fuel Composition Data	**Operating History Data**
Initial ^{235}U enrichment (wt %)	Reactor Type
Fuel assembly design	Specific Power (MW/MTU)
Presence of Integral Absorbers	Time since irradiation (cooling time)
Initial Fuel Composition	Burnup (MWd/MTU)
Fuel density	Coolant temperature/density
Fuel/Assembly Inhomogeneities	Fuel location in core

Generally, there is high confidence in defining a credible range of conditions for the initial state of the fuels and basic operating data for the primary reactor types (BWR, PWR, CANDU, MAGNOX, VVER, RBMK, graphite PRs). In order to look for trends or key isotopic ratios in these data, the uncertainty of the calculation method needs to be established through verification of the specific code, usually, by comparison with suitable and appropriate experiments. Commercial nuclear fuel vendor and some licensees have extensive experience in accurately calculating fuel performance and conditions for isotopes important to core reactivity and operations. For isotope important to accident safety, the commercial reactor calculations may be bounding values. For forensics applications, bounding calculated values may be of limited applicability as they wouldn't closely match the interdicted sample measurement.

Nuclear forensics can be used for both pre- and post-det (detonation) scenarios; however, the focus at PNNL is pre-detonation material forensics. A sample is analyzed to determine the major actinide isotopics, fission products, trace contaminants, and physical characteristics such as morphology. Once these are known, the following parameters can be looked at to trace the origin of the material:

- Signatures from ore body
- Solution residues
- Solid phase formation
- Reactor signatures
- Process impurities
- Metal conversion impurities
- Pathway signatures

[9] http://www.oecd-nea.org/science/wpncs/buc/

Analysis of isotopic ratios provides a means of identifying the neutron exposure history of pre-detonation materials. The transmutation of the isotopes and thus their ratios is dependent on the neutron flux spectrum and magnitude. Reactor signatures are the unique characteristics in the neutron flux that are inherent to the reactor type and operations. Within a given reactor core there is a range of neutron environments that provide a continuum of neutron spectra and magnitudes. As a result there is a natural spread in the isotopic ratios within each reactor core that is dependent on core location, operating reactor parameters and time.

Additional analyses of isotopic ratios can provide information that can further characterize the origin of the fuel within each reactor type. The ultimate objective of this work is to identify specific isotope ratios and provide detailed information about the fuel enrichment, burnup and cooling time for the specific sample. It is possible to determine additional details, such as the fuel assembly type, based on known data about these range and correlation of these parameters within a given reactor type. In the future, statistical methods can also be used to quantify a confidence level associated with these data.

4.0 Dataset Format

For the experimental data, values were first obtained from the SFCOMPO database then additional measurement data sources were found to expand the database. The measurement data was grouped by reactor type and the following header data were obtained:

- Reactor name
- Sample name
- Reactor type
- Fuel assembly dimensions
- Assembly name
- Pin number/ID
- Type of fuel
- Cooling time
- Measurement lab (experimental)
- Measurement Uncertainties
- Code system used (calculated)

These values were recorded in an Excel spreadsheet (see Figure 1), and each reactor system was included as a separate worksheet utilizing individualized tabs labeled with the name of the reactor and color-coded by reactor type (see Figure 2). The individual samples for a given reactor are represented by columns. Through the use of an Excel macro, all data within the dataset can be transferred to an Access database to facilitate data analysis.

The main content of each column contain the actual data by nuclide, burnup indicator and in some cases a ratio of nuclides and their uncertainties where available. The starting point for the list of nuclides to be included in SFCOMPO/NF was to be consistent with those nuclides identified as important to safety-related spent fuel applications in Table 4 of the *Spent Nuclear Fuel Assay Data for Isotopic Validation* report compiled by the EGADSNF[10] (hereafter referred to as the EGADSNF 2011 report). The final list as given in Table 2 includes all isotopes and isotope ratios that were reported for any measured reactor incorporated into SFCOMPO/NF. The list and order of nuclides are identical for all reactors (i.e. tabs) in the spreadsheet. For example (as seen in Figure 1) row 17 represents americium-241. Row 17 in any other reactor's tab will also be Am-241. The maintenance of consistent ordering is to facilitate preforming calculations on the values. This feature also made the transition to an Access database efficient.

Another feature of the spreadsheet format is that a column has been provided to include calculated values for each sample (as seen in column F of Figure 1). The inclusion of calculated values provides a basis for comparison of the measured values to a theoretical value for benchmarking and validation. More

[10] *Spent Nuclear Fuel Assay Data for Isotopic Validation, State-of-the-Art Report,* Nuclear Energy Agency Organization for Economic Co-operation and Development, NEA/NSC/WPNCS/DOC(2011) 5.

important to the application of these data for NF purposes is that the calculated values provide data that can be used to increase the number of isotopic ratios that can be compared. The level of detailed information that can be derived for an unknown sample is beyond simply the reactor type.

With the FY13 work, an additional column was added so that measurement and calculation uncertainties could be tabulated within the dataset (as seen in column E and G of Figure 1).

	A	B	C	D	E	F	G
1	Source:						
2	https://pnlweb.pnl.gov/projects/SFCOMPO/Shai	T. Yamamoto and M. Yamamoto, "Nuclear Analysis of PIE Data of Irradiated BWR 8x8-2 and 8x8-4					
3							
4	Completed source(s), uncertainties left off of values calculated from source(s)						
5	**Sample ID**			JPN2F2BWR-1			
6	**Reactor Type**			BWR			
7	**Fuel Assembly Dimensions**			8x8			
8	**Assembly**			2F2DN23			
9	**Pin**			B2			
10	**Axial Position (mm) (from bottom)**			39			
11	**Type**			UO2			
12	**Initial U-235 Enrichment**			0.71			
13	**Cooling Time (years)**			5.5			
14	**Measurement Lab/Code**			JAERI		JAERI	
15		**Units**		**Measured**	**Uncertainty**	**Calculated (E/C)**	**Uncertainty**
16	Ag-109	g/g U-238					
17	Am-241	kg/MTU initial		1.03E-02	2.00%		
18	Am-242	kg/MTU					
19	Am-242m	kg/MTU initial		7.98E-05	10.00%		
20	Am-242/Am-241						
21	Am-243	kg/MTU initial		5.84E-04	0.50%		
22	Am-243/Am-241			5.63E-02			

Figure 1. Spreadsheet Data Organization (Header)

54	Kr-83/Kr-86
55	Kr-83/Total Kr
56	Kr-84/Kr-83
57	Kr-84/Kr-86
58	Kr-84/Total Kr

JPDR | Tsuruga-1 | Fuk-Daiichi-3 | **Fuk-Daini-2** | Cooper | Monticello | Quad-Cities-1 | Fuk-Daini-2a | Obrigheim (19

Figure 2. Spreadsheet Data Organization (Reactor Tabs)

Table 2. List of Isotopes, Isotope Ratios and Burnup Values Included in SFCOMPO/NF
(All units are in kg/MTU unless otherwise noted.)

Ag-109[a]	Kr-86/Total Kr	**Ru-106** [b]
Am-241	**Mo-95** [a]	**Sb-125** [b]
Am-242	Nd-142 [b]	**Se-79** [b]
Am-242m	Nd-142/Total Nd	**Sm-147** [b]
Am-242/Am-241[b]	**Nd-143** [b]	Sm-148 [b]
Am-243	Nd-143/Total Nd	**Sm-149** [b]
Am-243/Am-241	Nd-144 [b]	**Sm-150** [b]
Burnup[c] **(U, Pu Isotope method)**	Nd-144/Total Nd	**Sm-151** [b]
Burnup (Cs-137 destructive)	**Nd-145** [b]	**Sm-152** [b]
Burnup (Cs-137 non-destructive)	Nd-145/Total Nd	Sm-154 [b]
Burnup (Nd-148 method)	Nd-146 [b]	Sn-126 [b]
Burnup (theoretical)	Nd-146/Total Nd	**Sr-90** [b]
Burnup	**Nd-148** [b]	**Tc-99** [b]
Ce-144 [b]	Nd-148/Total Nd	Total Pu [b]
Cm-242	Nd-148/U	Total Pu + U
Cm-243 [b]	Nd-148/U-238	Total Pu/Total U
Cm-244	Nd-150 [b]	Total Pu/Total U (atoms)
Cm-245 [b]	Nd-150/Total Nd	Total U
Cm-246 [b]	**Np-237** [b]	U-232 [b]
Cm-247 [b]	Pd-107	**U-234** [b]
Cs-133	**Pm-147**	U-234/Total U
Cs-134	Pu-236	**U-235**
Cs-134/Cs-137 (activity)	**Pu-238**	U-235 Depletion
Cs-135 [b]	Pu-238/239	U-235/Total U
Cs-137	Pu-238/Total Pu	U-235/Total U (atoms)
Cs-137/U-238	Pu-238/Total Pu (atoms)	U-235/Total U-initial (atoms)
Eu-151 [a]	**Pu-239**	U-235/U-238
Eu-153 [a]	Pu-239/Total Pu	**U-236** Build up
Eu-154 [b]	Pu-239/Total Pu (atoms)	U-236/Total U
Eu-155 [a]	Pu-239/U-238	U-236/Total U (atoms)
Eu-154/Cs-137 (activity)	**Pu-240**	U-236/U-238
Gd-154	Pu-240/Pu-239	**U-238**
Gd-155	Pu-240/Total Pu	U-238 Depletion
Gd-156	Pu-240/Total Pu (atoms)	U-238/Total U
Gd-157	**Pu-241**	U-238/Total U (atoms)
Gd-158	Pu-241/Pu-239	Xe-131/Total Xe
Gd-160	Pu-241/Total Pu	Xe-131/Xe-134
I-129	Pu-241/Total Pu (atoms)	Xe-132/Total Xe
Kr-83/Kr-86	**Pu-242**	Xe-132/Xe-131
Kr-83/Total Kr	Pu-242/Pu-239	Xe-132/Xe-134
Kr-84/Kr-83	Pu-242/Total Pu	Xe-134/Total Xe
Kr-84/Kr-86	Pu-242/Total Pu (atoms)	Xe-136/Total Xe
Kr-84/Total Kr	**Rh-103** [a]	Xe-136/Xe-134
Kr-85/Kr-86	**Ru-101** [a]	**La-139**

[a] Units are g/g U-238 [b] Units are kg/MTU initial [c] All burnup units are GWd/MTU

Isotopes listed in **BOLD** are identified as commonly measured nuclides of importance for safety-related spent fuel applications in the EGADSNF 2011 report.

5.0 Measured Data

The measured data from SFCOMPO were used as a starting point to baseline SFCOMPO/NF. Over FY11 and FY12, additional data were added to increase the data available for comparison. New measurement data were extracted from the following eight documents:

PWR – TMI, Turkey Point and Gösgen

- SCALE 5.1 Predictions of PWR Spent Nuclear Fuel Isotopic Compositions, Oak Ridge National Laboratory (Radulescu 2010)

PWR – TMI and BWR – Quad Cities

- Analysis of Spent Fuel Nuclear Samples from Three Mile Island and the Quad Cities Reactors: Final Repot, Argonne National Laboratory (Wolf 2000)

BWR – Fuk-Daini2

- Compilation of Measurement and Analysis Results of Isotopic Inventories of Spent BWR Fuels (Yamamoto 2009)

Graphite Reactor – B Reactor, K West Reactor and C Reactor

- Experimental Isotopic Analysis of Point Exposure Data in Hanford Production Reactor Fuels, Douglas United Nuclear Inc. (Toffer 1970)

CANDU – NPD, Bruce A and Pickering

- Verification and Validation of the ORIGEN-S Code and Nuclear Data Libraries, Atomic Energy of Canada Limited. (Gauld 1995)

MAGNOX – Hunterston A, Bradwell

- FISPIN10 Validation Review, British Nuclear Fuels. (Parker 2001)

VVER – Novovoronezh NPP-4

- Radiochemical Assays of Irradiated VVER-440 Fuel for Use in Spent Fuel Burnup Credit Activities, Lawrence Livermore National Laboratory. (Jardine 2005)

VVER – Novovoronezh NPP-3, NPP-4, NPP-5, Kalinin, Balakovo and RBMK- Leningrad

- Destructive Analysis of the Nuclide Composition of Spent Fuel of WWER-440, WWER-1000 and RBMK-1000 Reactors, Khlopin Radium Institute. (Makarova 2008)

As of FY13, all sources are included and linked to their respective data within the Excel dataset for ease of reference.

The new measurements were incorporated to expand the range of operating parameters in SFCOMPO/NF for the PWR and BWRs. The operating parameter ranges for the seven reactor types currently documented in SFCOMPO/NF are summarized in Table 3 below and represent a large range in operating parameters for the reactor types shown. Data for graphite PRs were also added to provide a reference reactor type that could be used to identify reactor types. It is well established that the operating conditions and therefore isotopic composition of used fuel from PRs would be highly distinguishable from the LWR data.

Table 3. Range of Operating Parameters in Current SFCOMPO/NF by Reactor Type

Reactor Type	Enrichment (wt%U-235)	Burnup (GWd/MTU)	Cooling time* (years)
PWR	2.453 - 5.07	0.00891 – 55.7	0 – 10.8
BWR	0.71 – 4.5	2.21 – 59.1	0 – 6.7
Graphite PR	0.143 – 2.1	0.259 – 2.083	0.107 – 0.249
CANDU	0.71	6.2	0.499 – 15.315
MAGNOX	0.71	3.90 - 8.93	0.704 – 1.74
VVER	3.3 – 4.4	8.7 – 51.7	3.0 – 10.4
RBMK	1.8 – 2.09	6.2 – 27.7	0.80 – 4.7

*The cooling times of 0 listed above have likely been mathematically altered to obtain this value. Samples are not typically analyzed within seconds or even hours of removal from a reactor.

6.0 Addition of Model Data

In addition to the new measurements that were added, calculated data was also incorporated into SFCOMPO/NF. The calculated data was in the form of E/C (experimental values divided by calculated values) as they were obtained from the original references. The calculated values were placed into the spreadsheet in columns next to the measured experimental values for ease of comparison.

The primary driver to include additional calculated data in SFCOMPO/NF (aside from validation of models) is that not all historical samples include all or even most of the nuclides identified in Table 2. In order to provide a larger range of information to be evaluated for signatures, calculated data have been incorporated. SFCOMPO/NF includes calculated data consistent with a subset of the measured data.

The calculated values added to the SFCOMPO/NF dataset were included from two references:

- SCALE 5.1 Predictions of PWR Spent Nuclear Fuel Isotopic Compositions (Radulescu 2010)
- FY11 Summary Report: Plutonium Signatures (Brady Raap 2012)

7.0 Comparison of Actinide Isotope Ratios

With all of the measured data compiled into a spreadsheet, the ratios of the plutonium isotopes could be compared for the different reactor types. There are five plutonium isotopes of interest that are generated through various decay and neutron capture reactions: Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242. Since Pu-239 is the most prevalent, it is standard practice to ratio the other plutonium isotopes to Pu-239.

The following graphs compare measured data by reactor type in relation to their plutonium ratios. Some of these ratio graphs were presented in last year's summary report[11]. The Pu-240/Pu-239 ratio is a reasonable indicator of reactor burnup with the PWR data shown in Figure 3. The different reactor types would show a similar trend, so the ratio will be the x-axis for comparison to the other ratios. Figure 4 shows the Pu-238/Pu-239 ratio versus the Pu-240/Pu-239 burnup indicator with uncertainties where available. This plot shows that the spread in the data is much larger than the uncertainty measurements.

Figure 5 – Figure 10 show each reactor's data fitted with a simple second order least squares fit with uncertainty bands optimized to encompass ≥60% of the data. There is not a separate plot for the CANDU reactor as there was only one data point for Pu-238. Also, for the PWR data, shown in Figure 5, the TMI data was plotted but not included in the fit as the samples had unconventionally high burnups. For the VVER data shown in Figure 8, the extreme outlier was excluded from the fit.

Figure 11 shows a comparison of the PWR, BWR, and RBMK data, fits, and uncertainty bands. Figure 12 shows the Pu-241/Pu-239 ratio, with Figure 13 – Figure 18 showing each reactor's data fitted with the same technique and uncertainty bands, with the exception of CANDU since this reactor type only had 3 data points for Pu-241. Figure 19 shows a comparison of the PWR, BWR, and RBMK data, fits, and uncertainty bands. Figure 20 shows the Pu-242/Pu-239 ratio, with Figure 21 – Figure 26 showing each reactor's data fitted with the same technique and uncertainty bands, with the exception of CANDU since this reactor type only had 3 data points for Pu-242. Figure 27 shows a comparison of the PWR, BWR, and RBMK data, fits, and uncertainty bands.

This data comparison highlights the overlap in data giving a probability that a specific ratio comparison can be identified as a specific reactor. For this analysis, only plutonium isotopic ratios were used to compare the different reactor types. Other isotopes including actinides and fission products may allow for better identification of reactor type.

When plotting the plutonium ratios general trends can be identified in the data for different reactor types. The graphite reactor data has a much different slope than the light water reactors for the Pu-238/Pu-239 and Pu-242/Pu-239 ratios. While there is significant overlap in the PWR and BWR reactor data, the mean trend though the data shows a different slope in the plutonium isotopic ratios for these two types of LWRs. In general, the PWR and VVER data follow the same trend since they are both light water cooled and pressurized water reactors. The Graphite PRs and RBMK reactors also follow the same trend as they are both graphite moderated reactors.

[11] Brady Raap MC, BA Collins and CJ Francy. 2012. *FY11 Summary Report on the Augmentation of the Spent Fuel Composition Database for Nuclear Forensics: SFCOMPO/NF*. PNNL-21939, Pacific Northwest National Laboratory, Richland, WA.

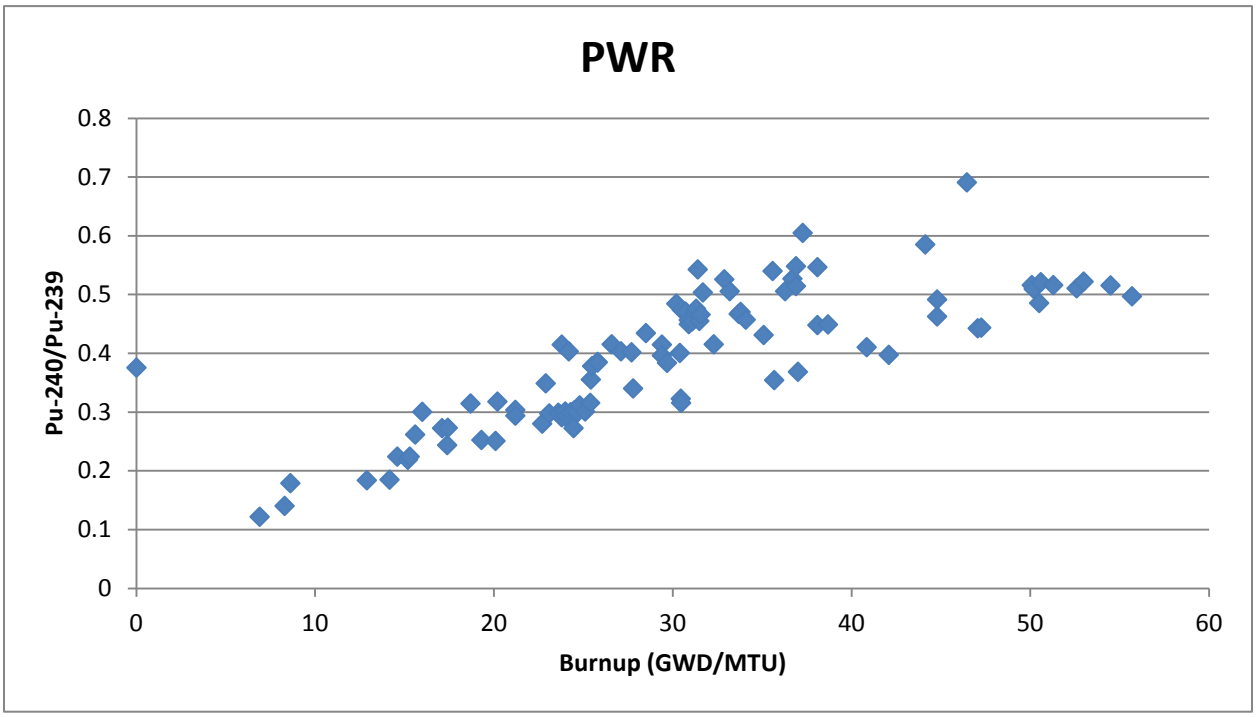

Figure 3 Pu-240/Pu-239 Plotted Against Burnup for PWR Measurement Data

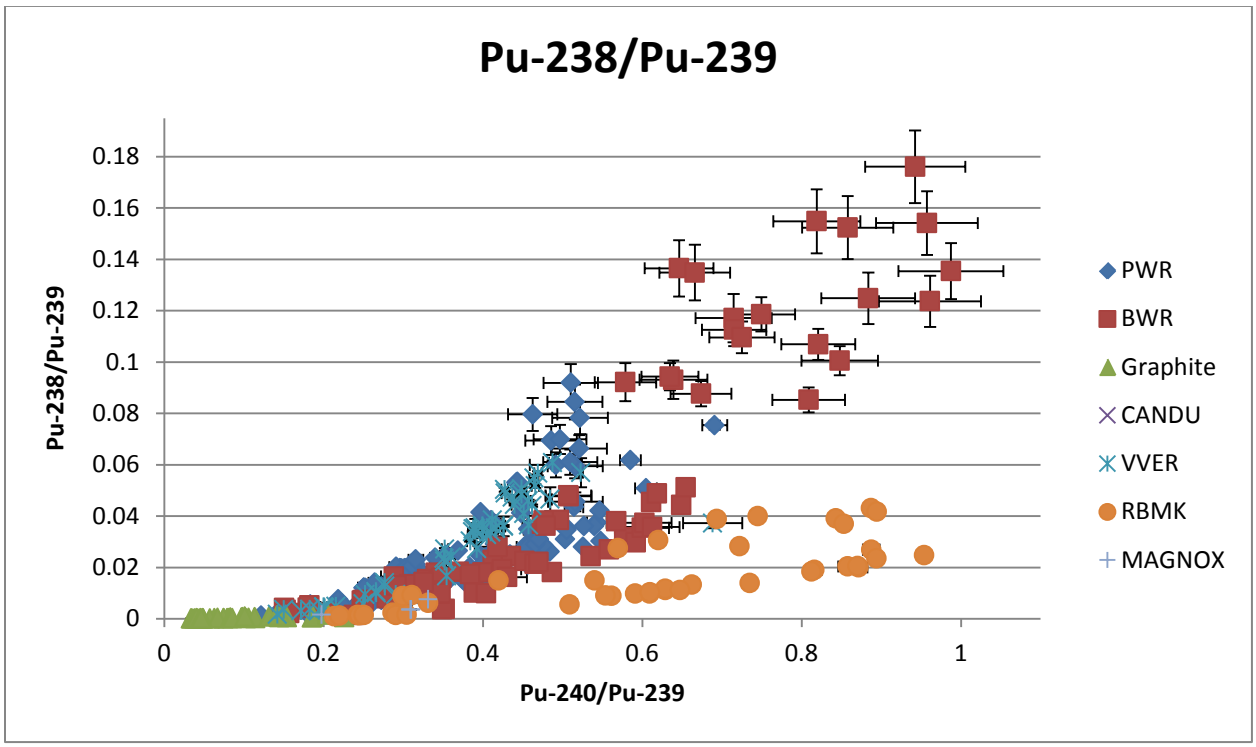

Figure 4 Comparison of Pu-238/Pu239 Ratios by Reactor Type with Error Bars where Available

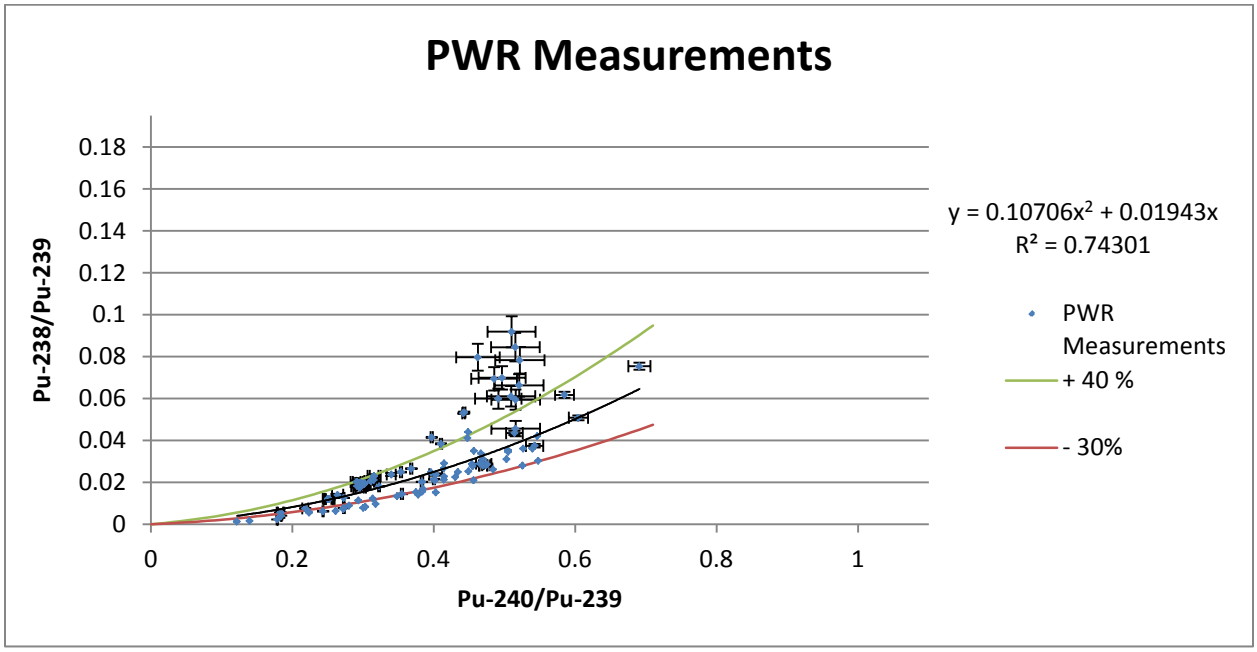

Figure 5 PWR Pu-238/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with +40% and -30% Uncertainty Bands

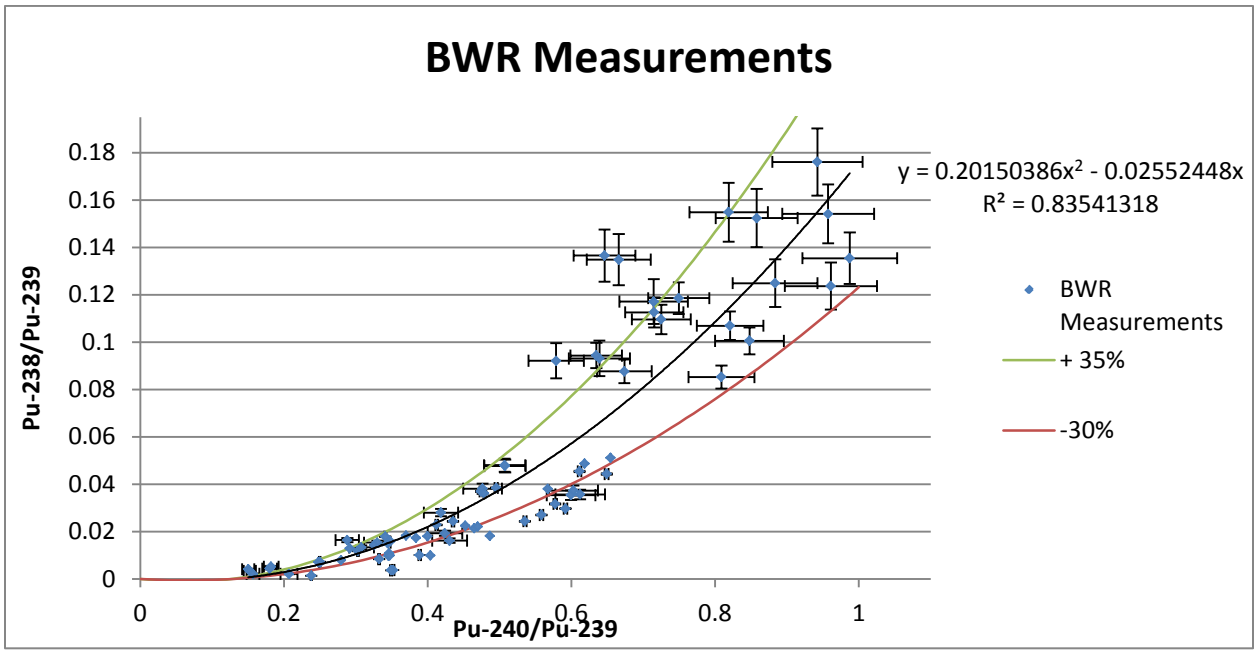

Figure 6 BWR Pu-238/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +35% and -30% Uncertainty Bands

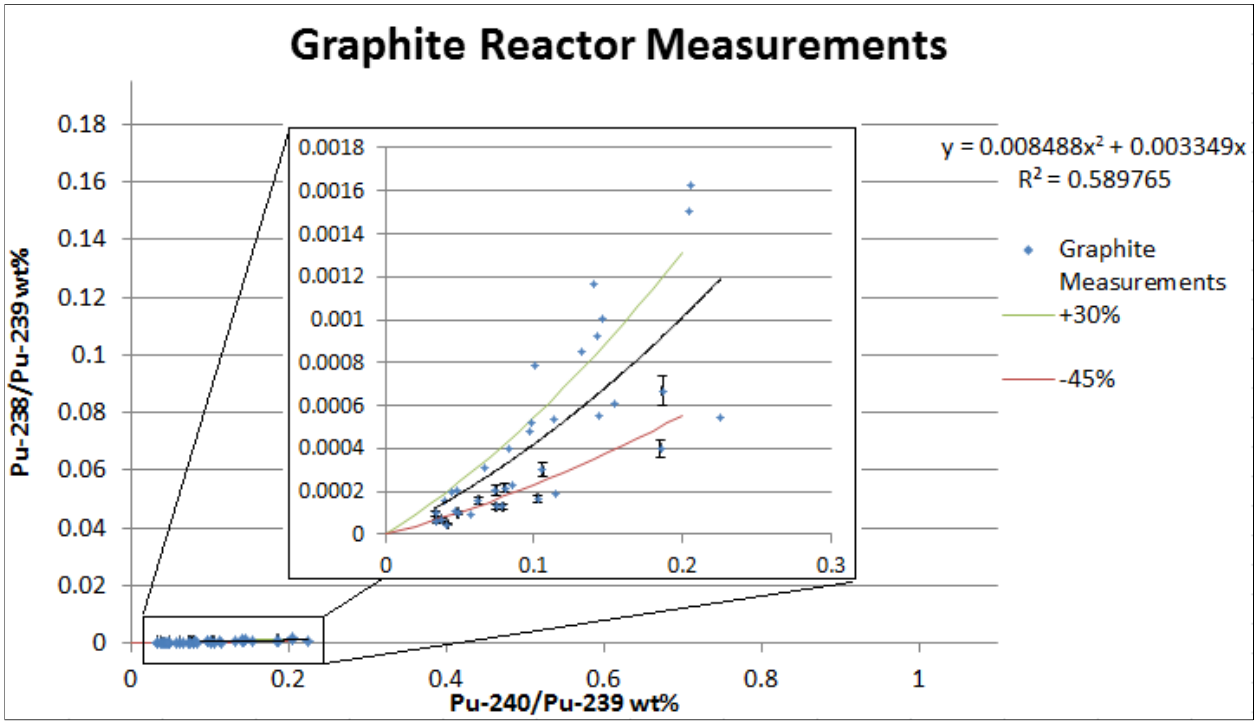

Figure 7 Graphite Reactor Pu-238/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +30% and -45% Uncertainty Bands

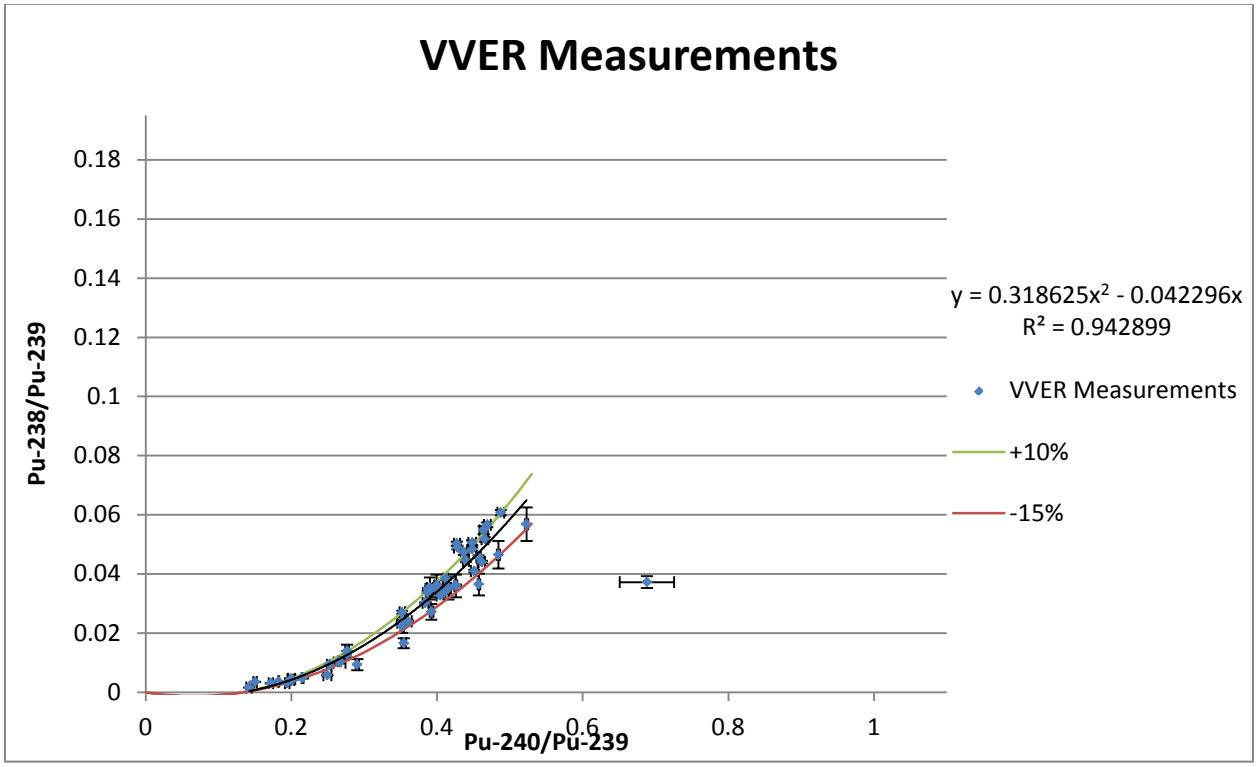

Figure 8 VVER Pu-238/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +10% and -15% Uncertainty Bands

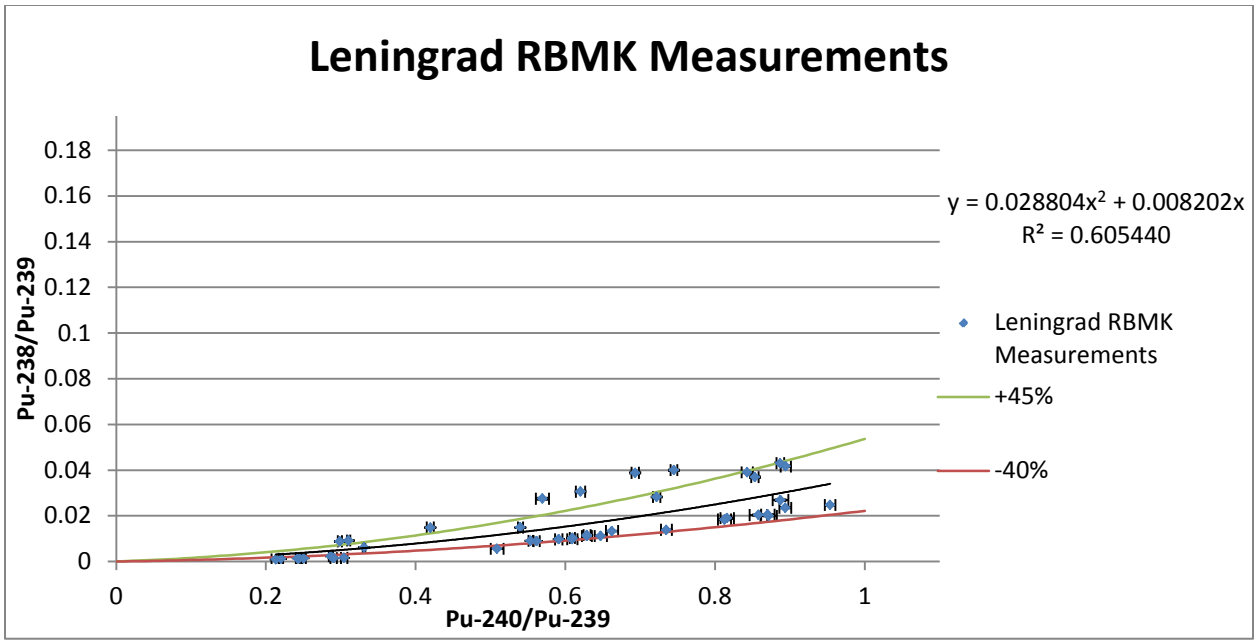

Figure 9 Leningrad RBMK Pu-238/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +45% and -40% Uncertainty Bands

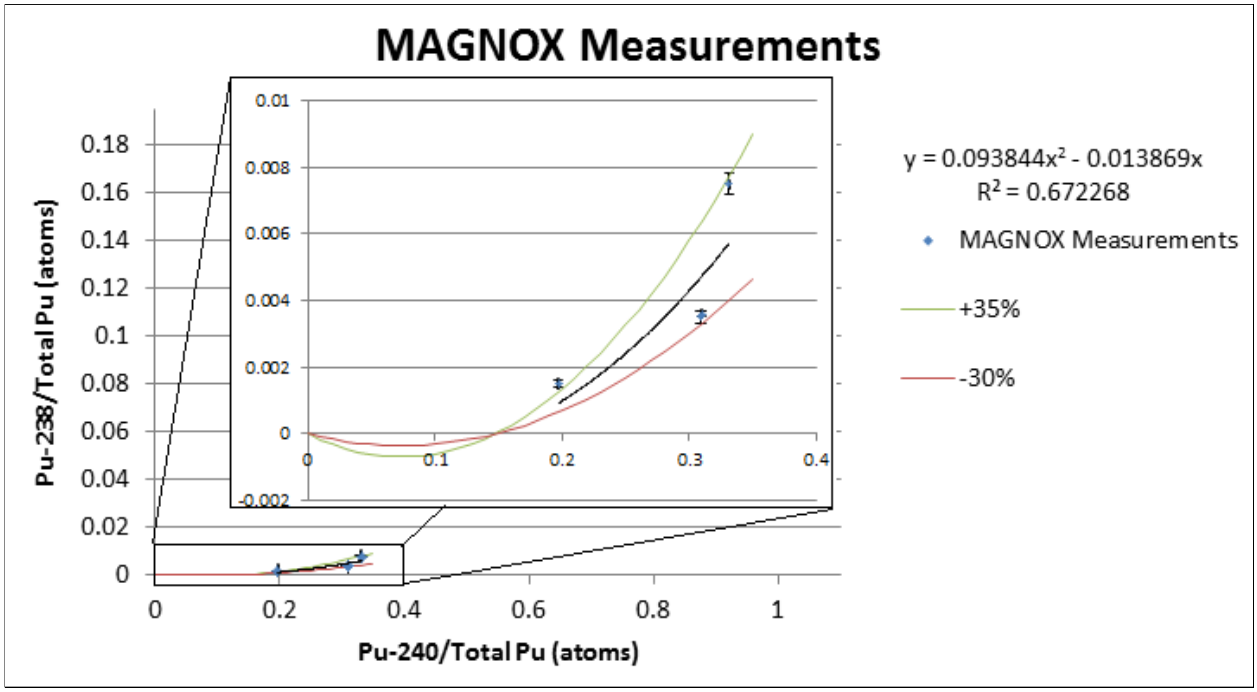

Figure 10 MAGNOX Pu-238/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +35% and -30% Uncertainty Bands

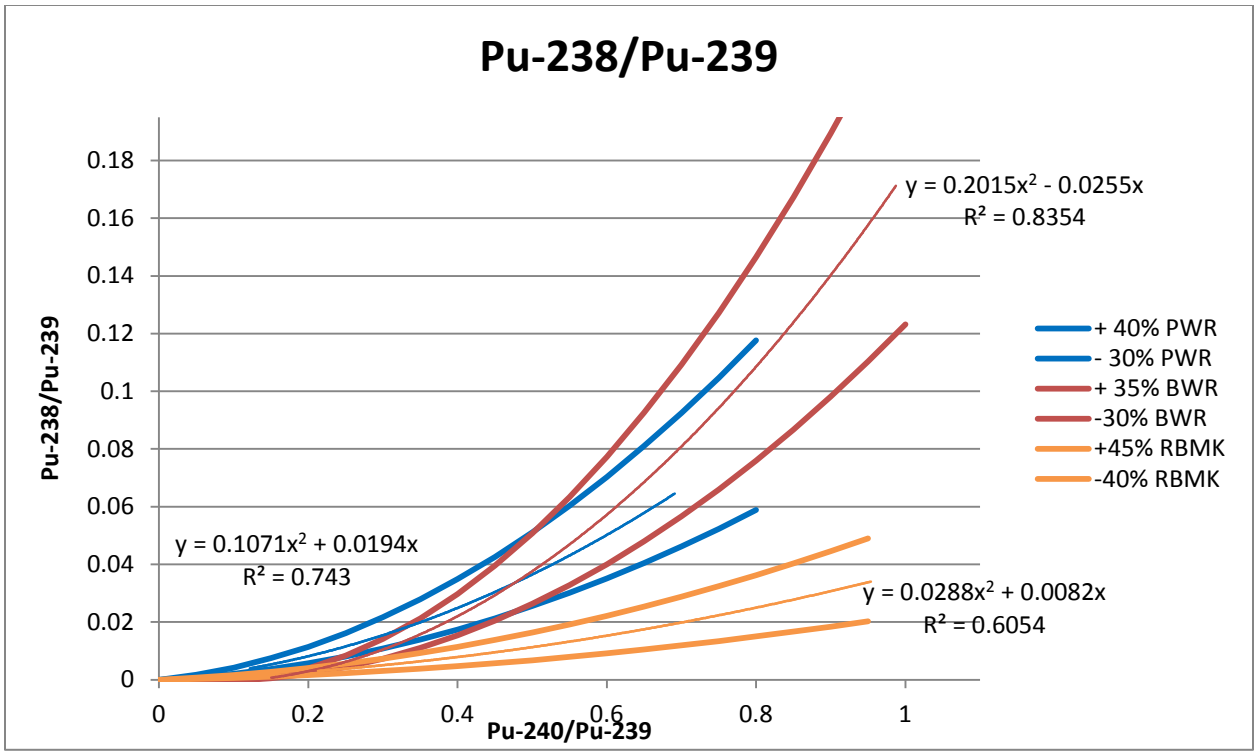

Figure 11 PWR, BWR, and RBMK Pu-238/Pu-239 Measurement Data Comparison with Second Order Least Squares Fits and Uncertainty Bands

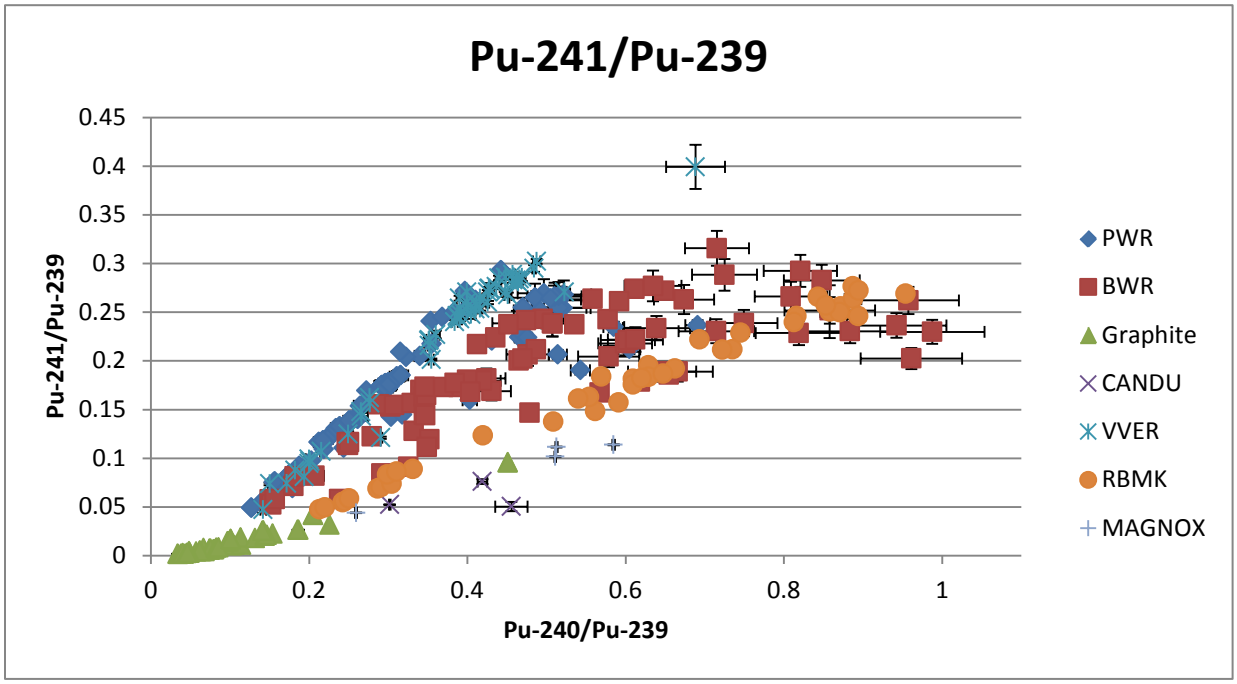

Figure 12. Comparison of Pu-241/Pu-239 Ratios by Reactor Type with Error Bars where Available

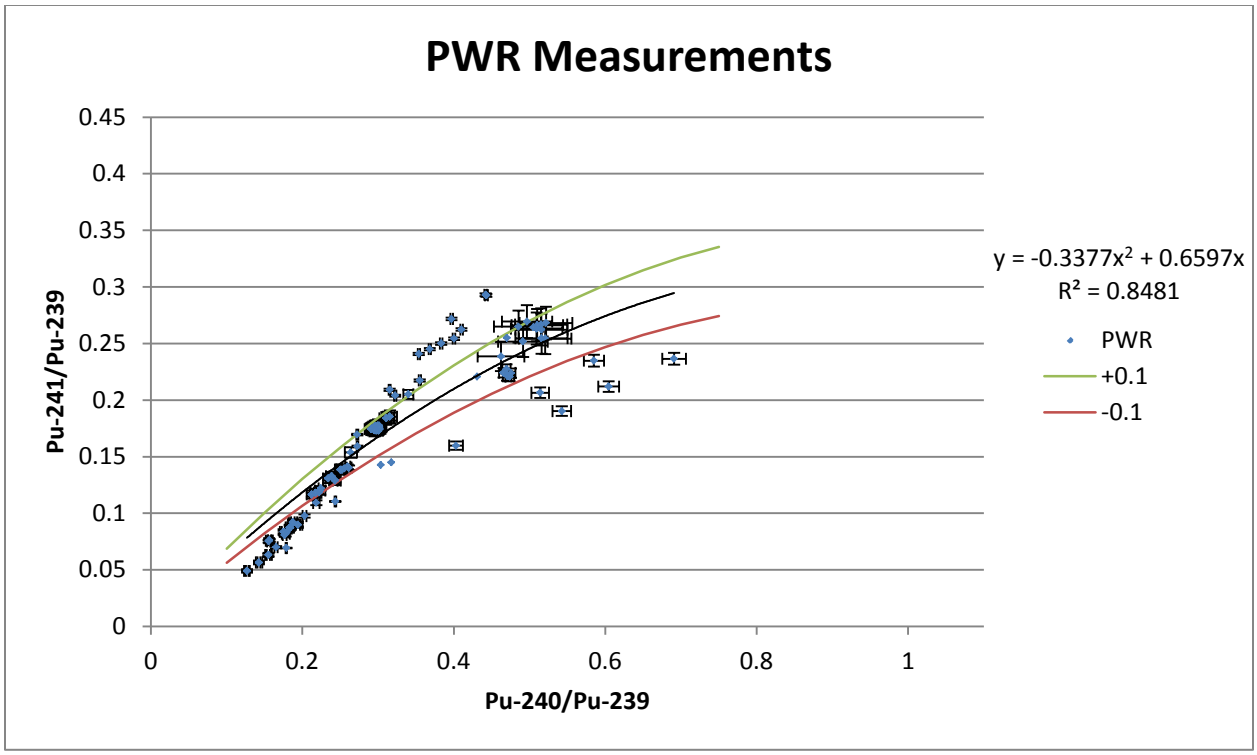

Figure 13 PWR Pu-241/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +10% and -10% Uncertainty Bands

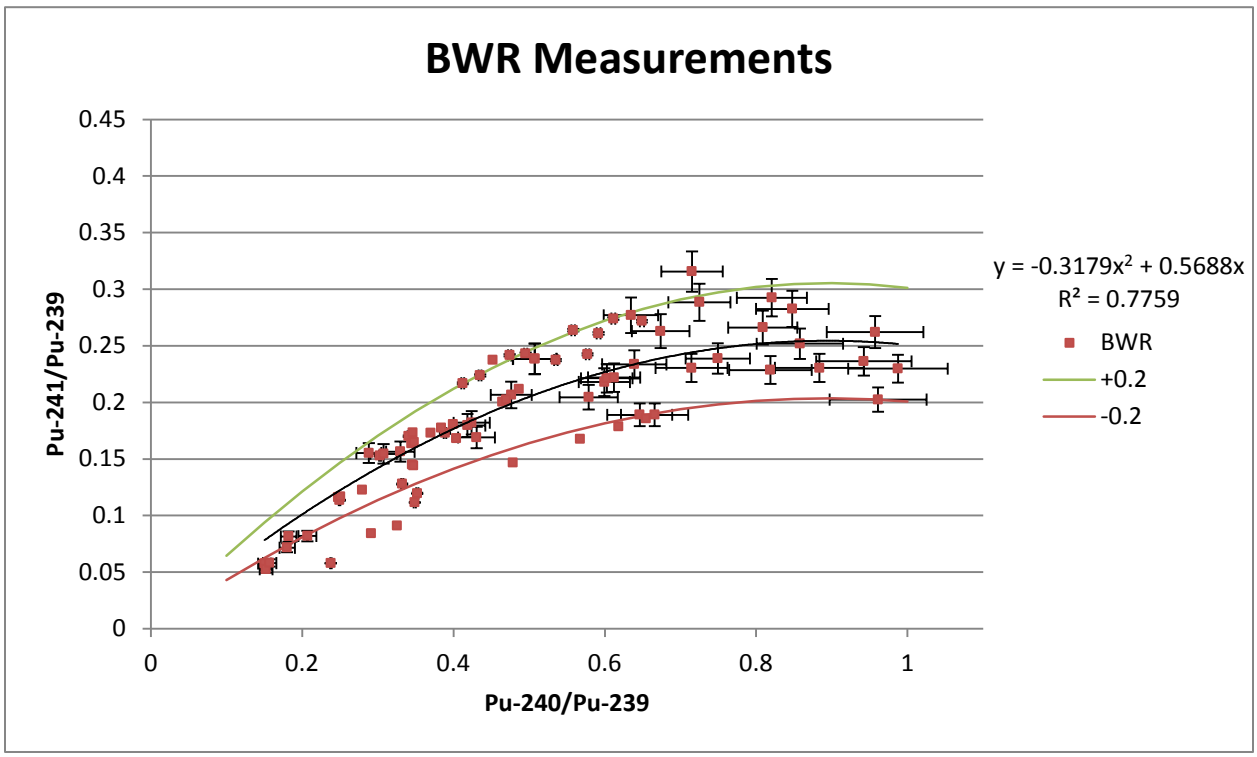

Figure 14 BWR Pu-241/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +20% and -20% Uncertainty Bands

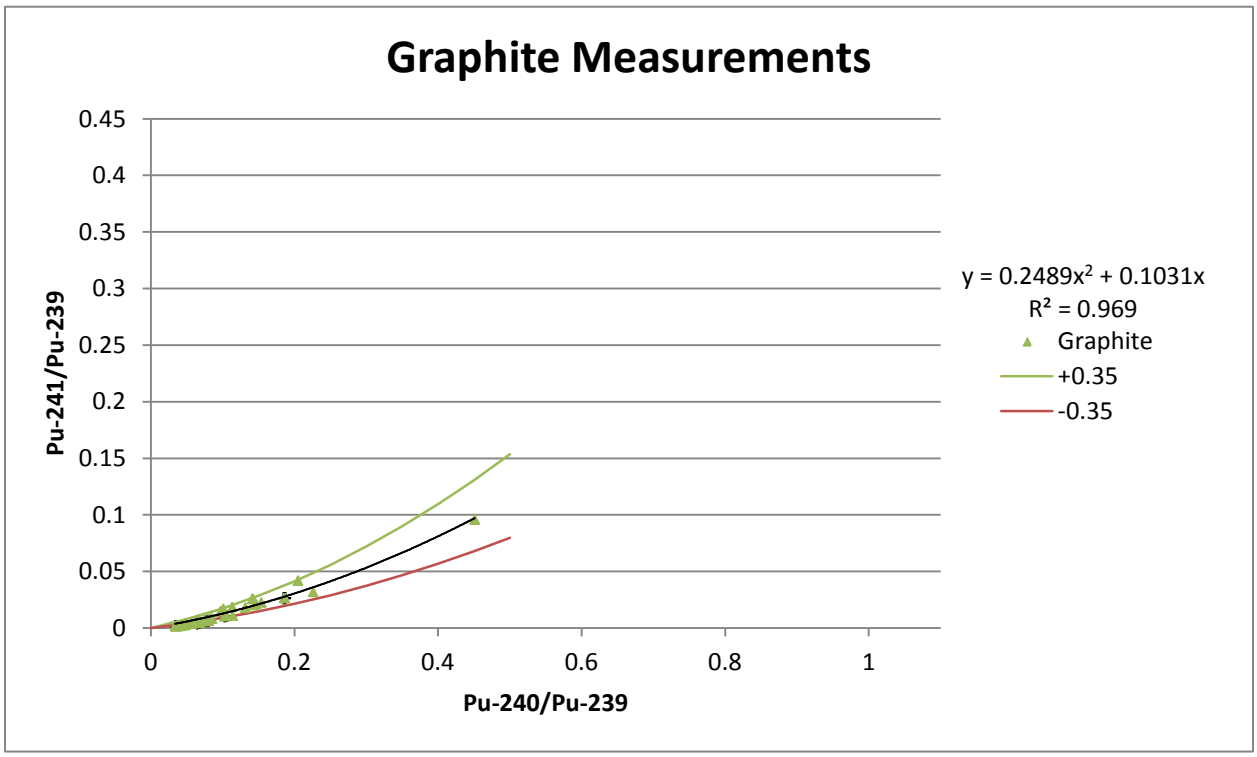

Figure 15 Graphite Reactor Pu-241/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +35% and -30% Uncertainty Bands

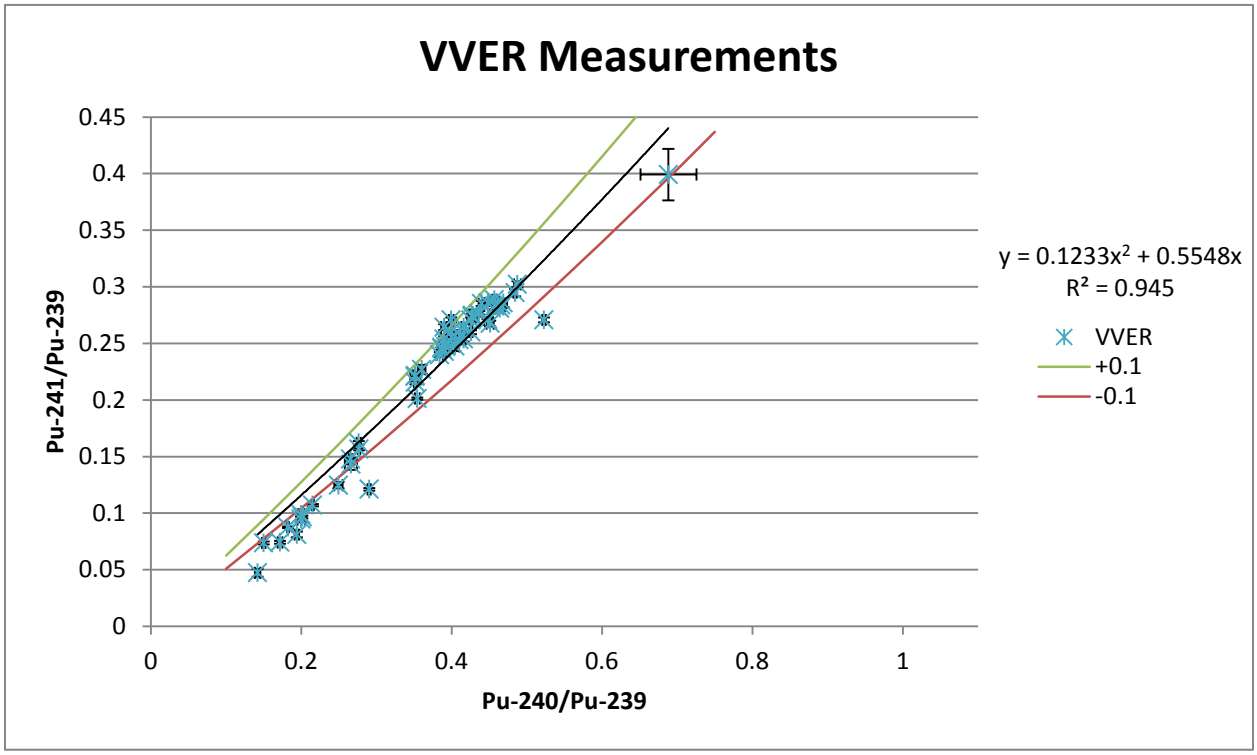

Figure 16 VVER Pu-241/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +10% and -10% Uncertainty Bands

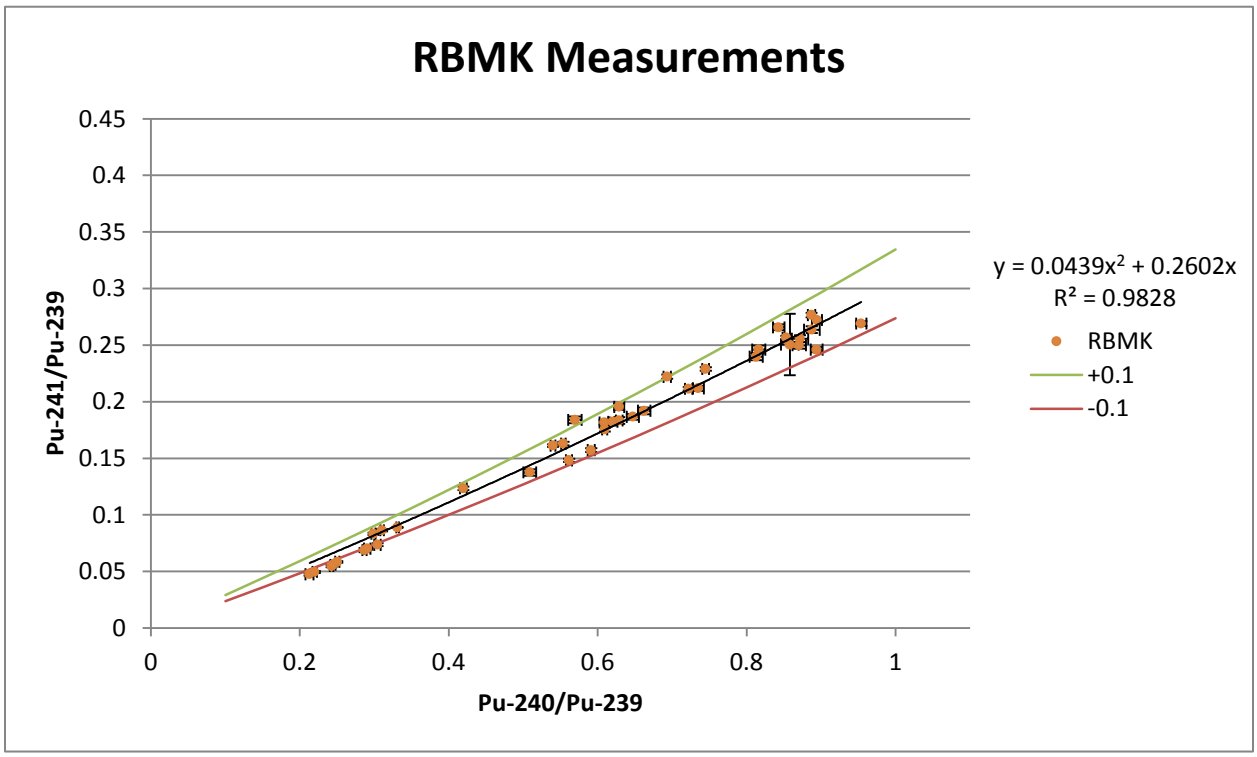

Figure 17 RBMK Pu-241/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +10% and -10% Uncertainty Bands

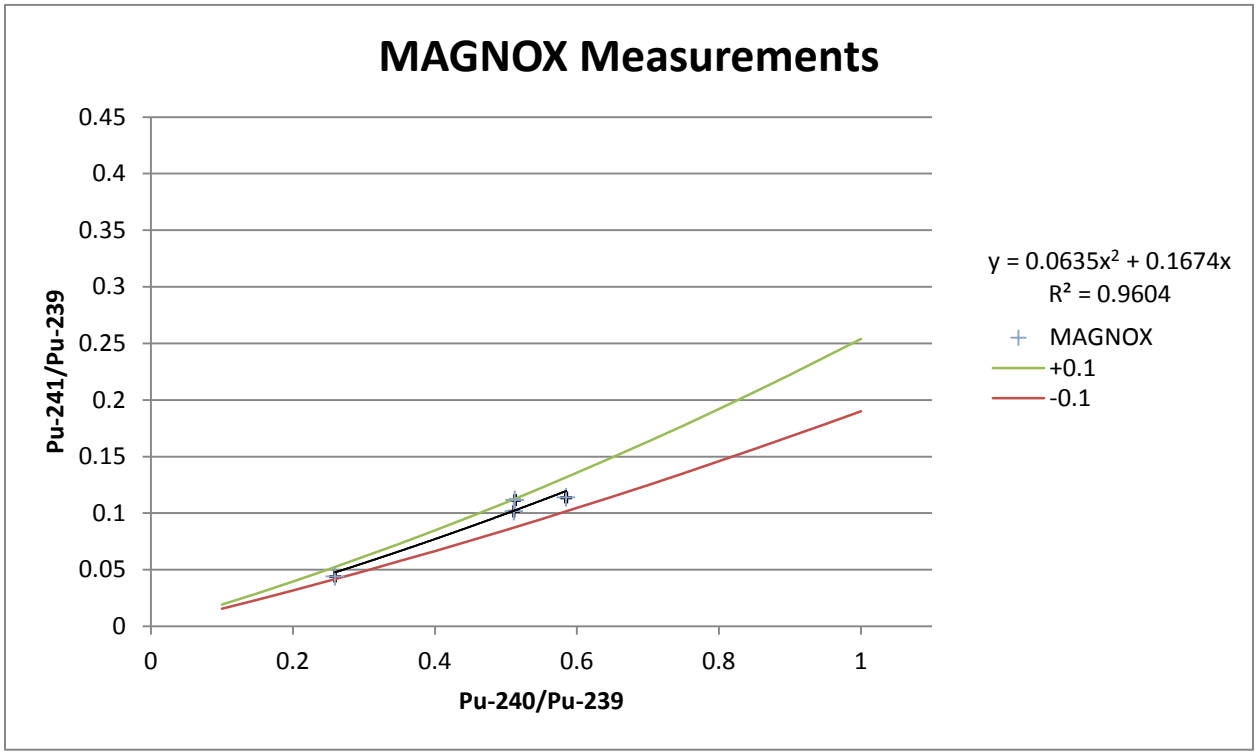

Figure 18 MAGNOX Pu-241/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +10% and -10% Uncertainty Bands

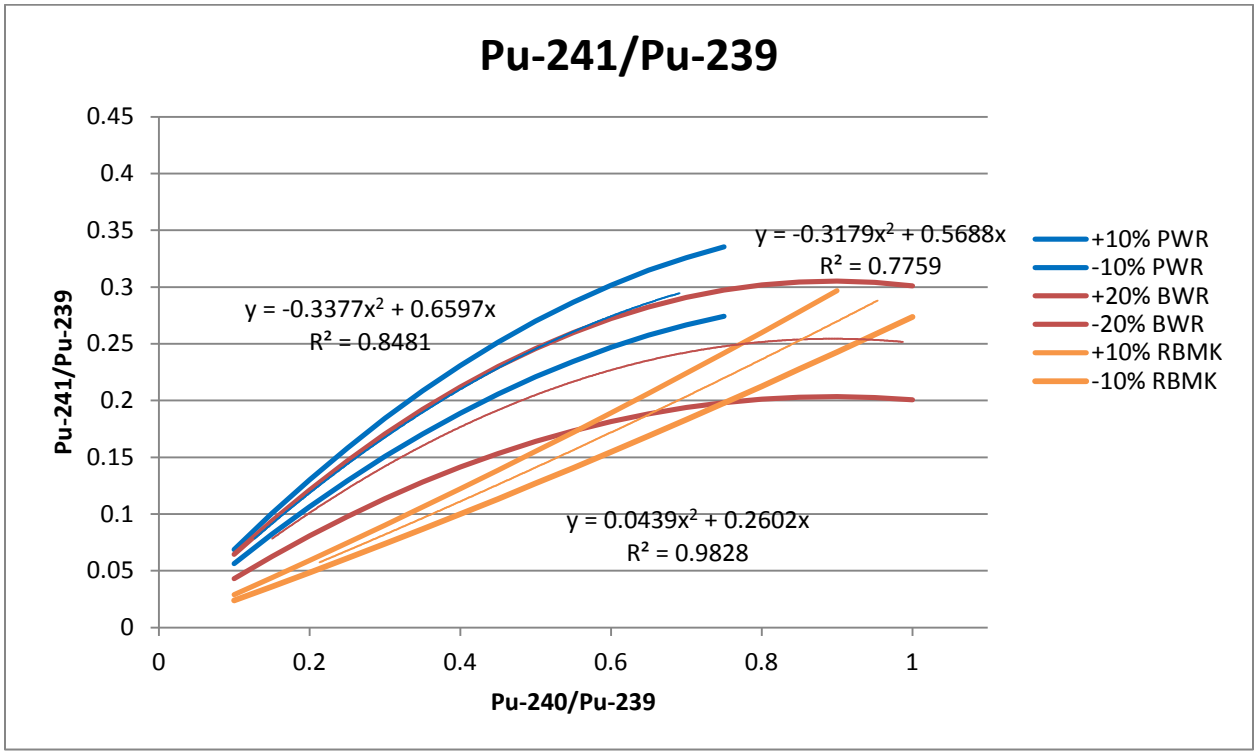

Figure 19 PWR, BWR, and RBMK Pu-241/Pu-239 Measurement Data Comparison with Second Order Least Squares Fits and Uncertainty Bands

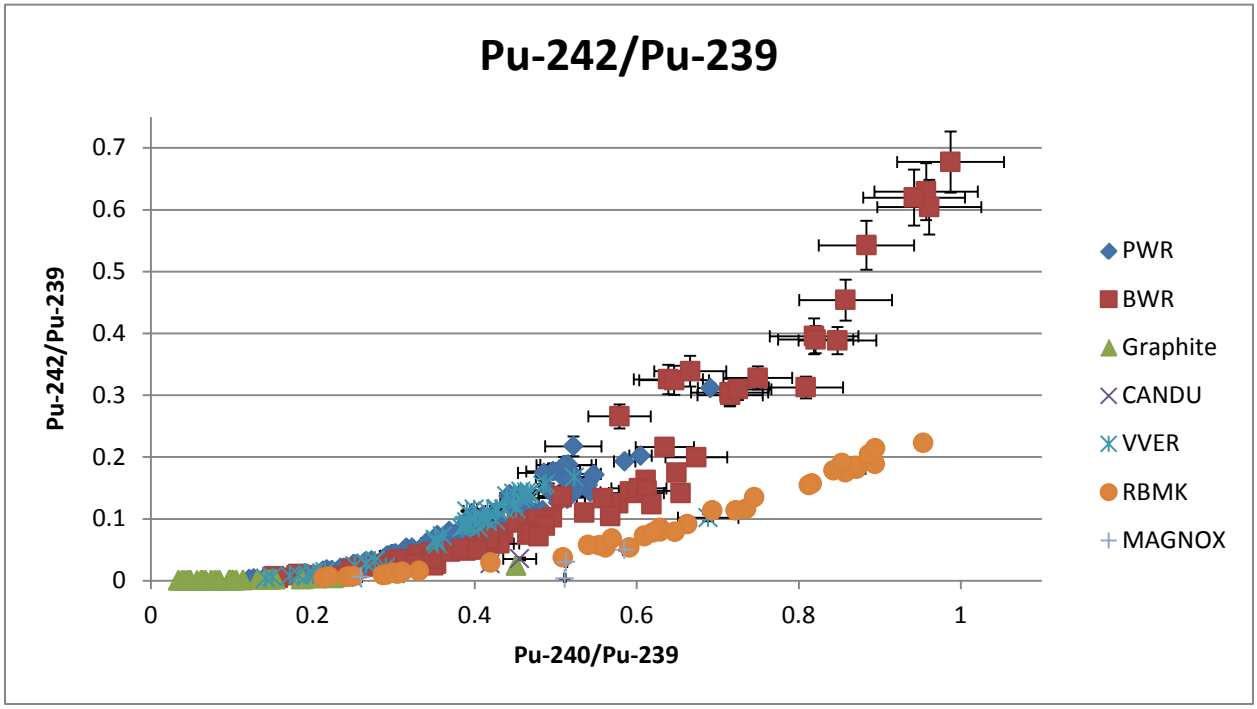

Figure 20. Comparison of Pu-242/Pu-239 Ratios by Reactor Type with Error Bars where Available

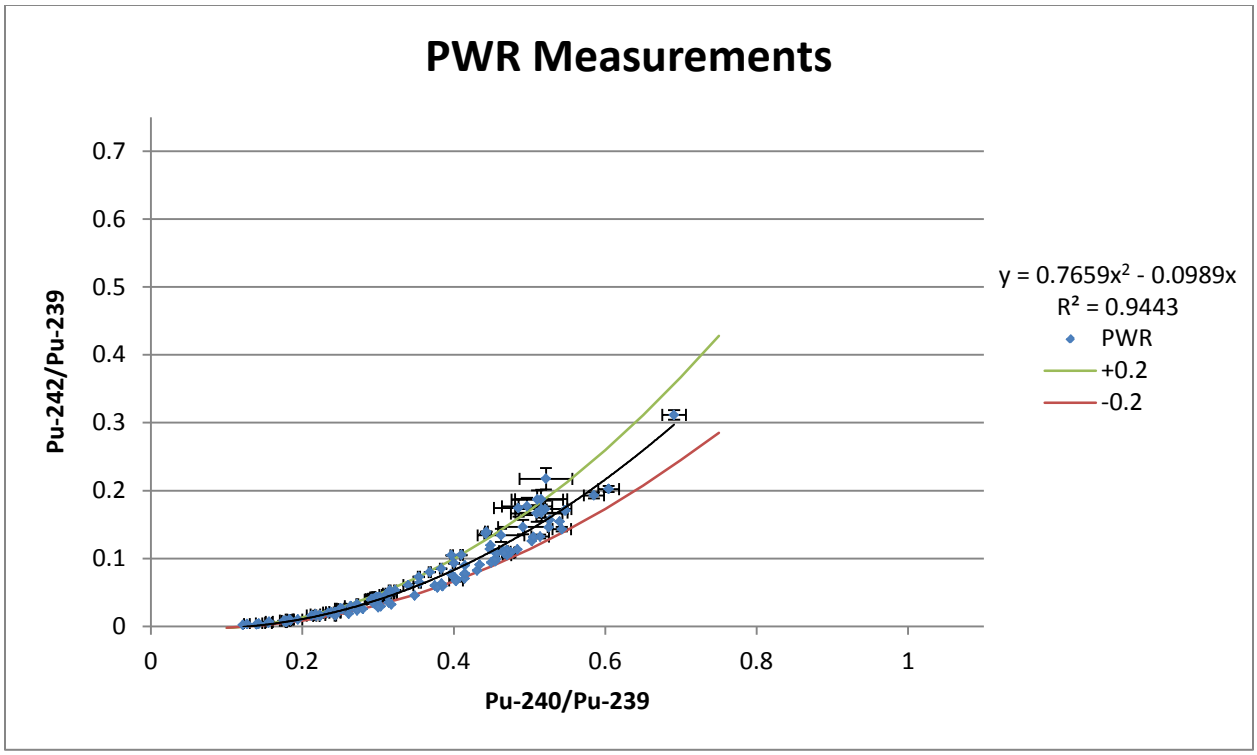

Figure 21 PWR Pu-242/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +20% and -20% Uncertainty Bands

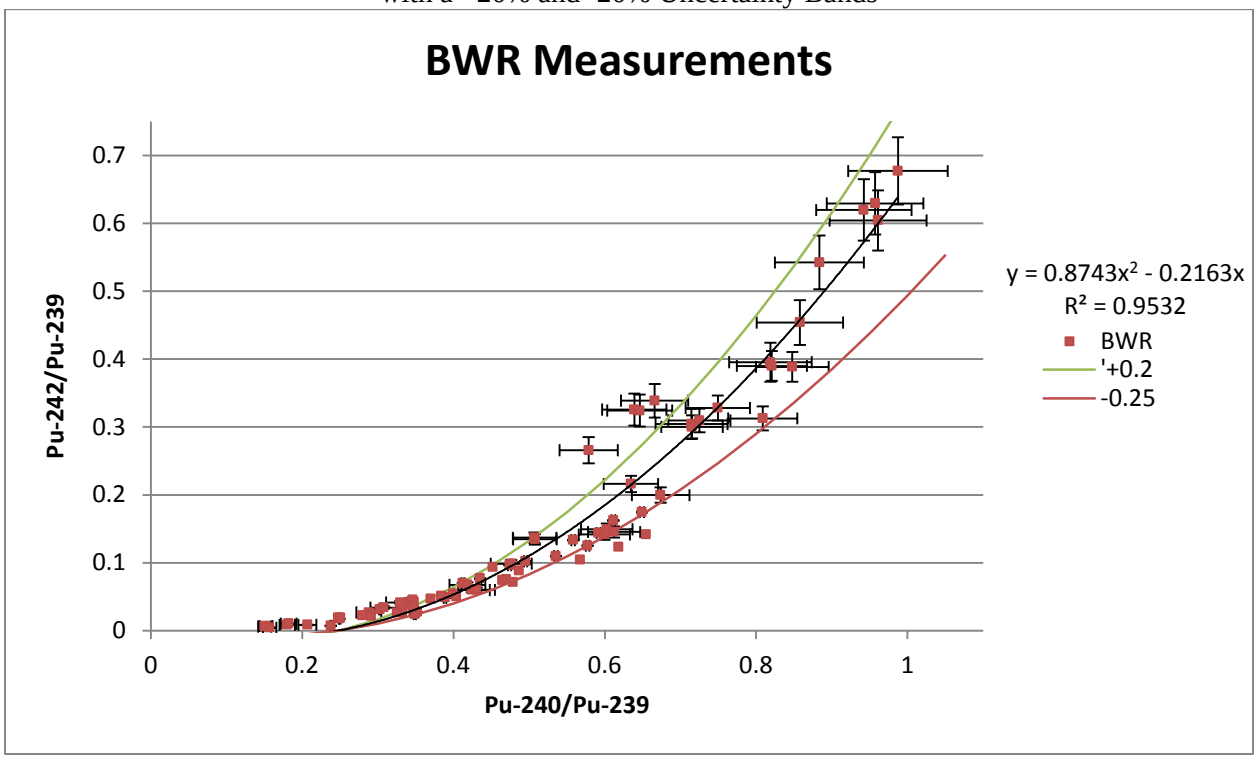

Figure 22 BWR Pu-242/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +20% and -25% Uncertainty Bands

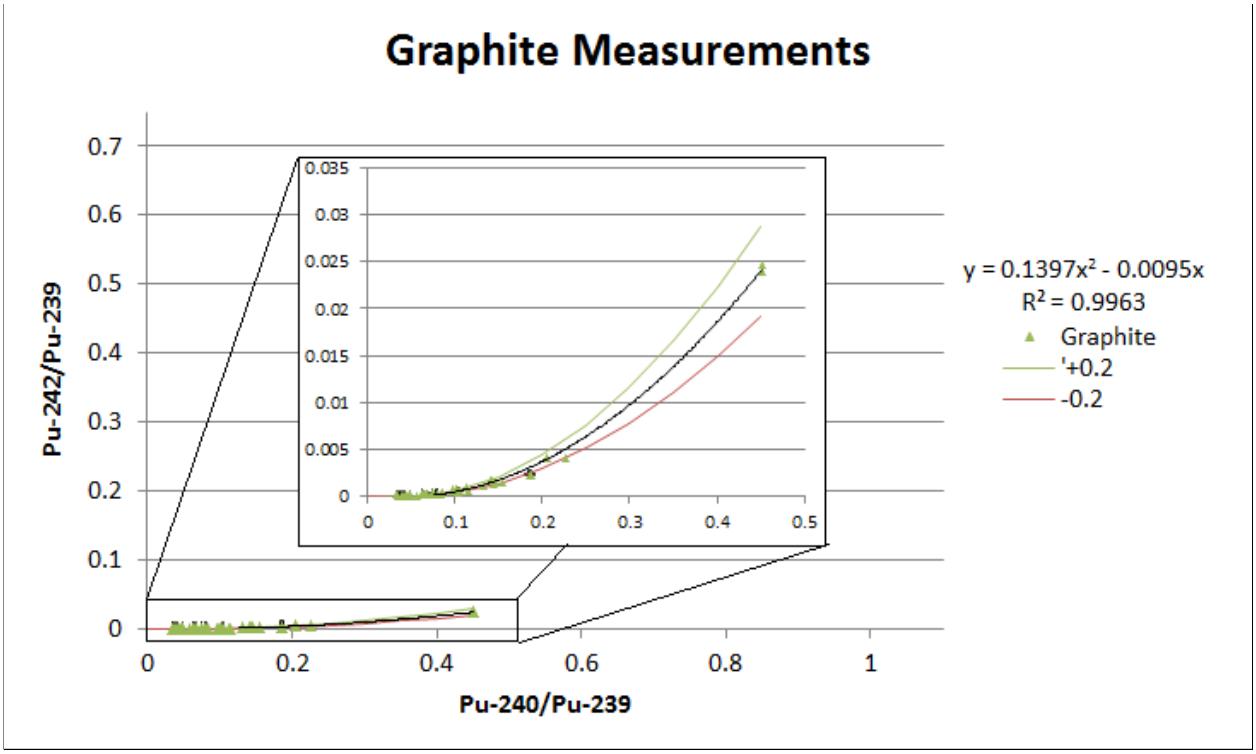

Figure 23 Graphite Reactor Pu-242/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +20% and -20% Uncertainty Bands

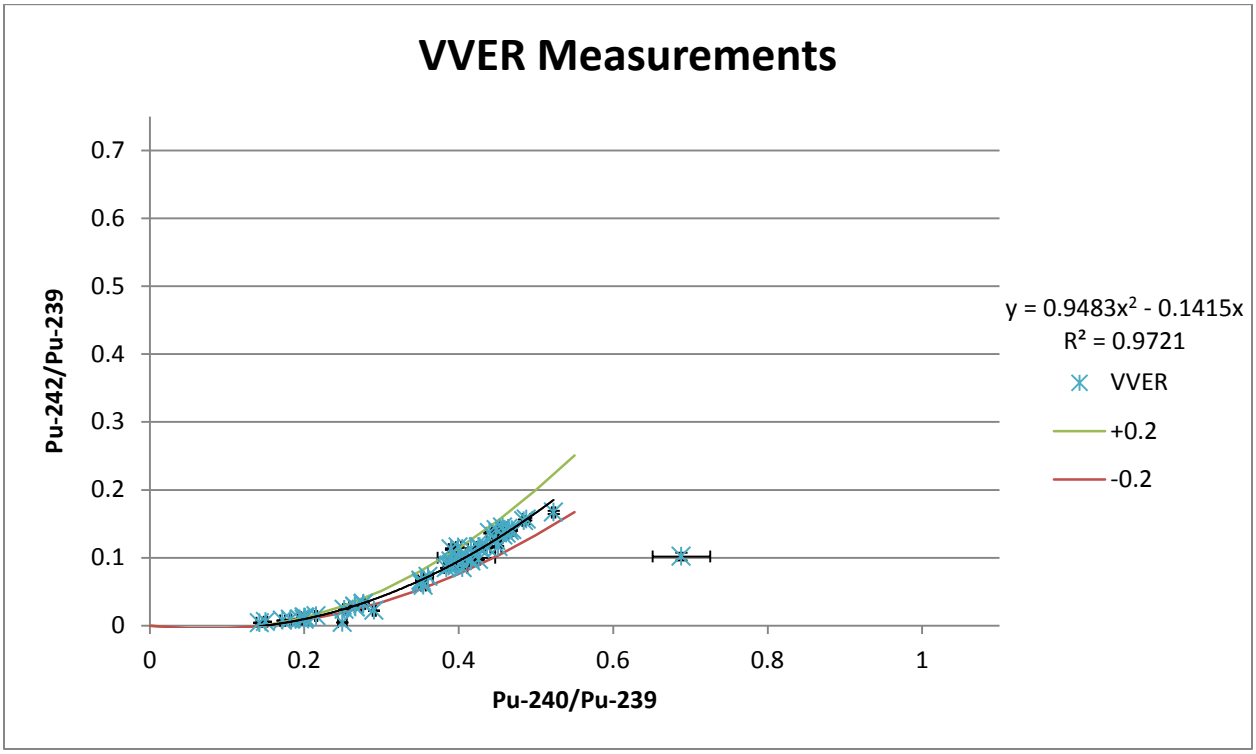

Figure 24 VVER Pu-242/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +20% and -20% Uncertainty Bands

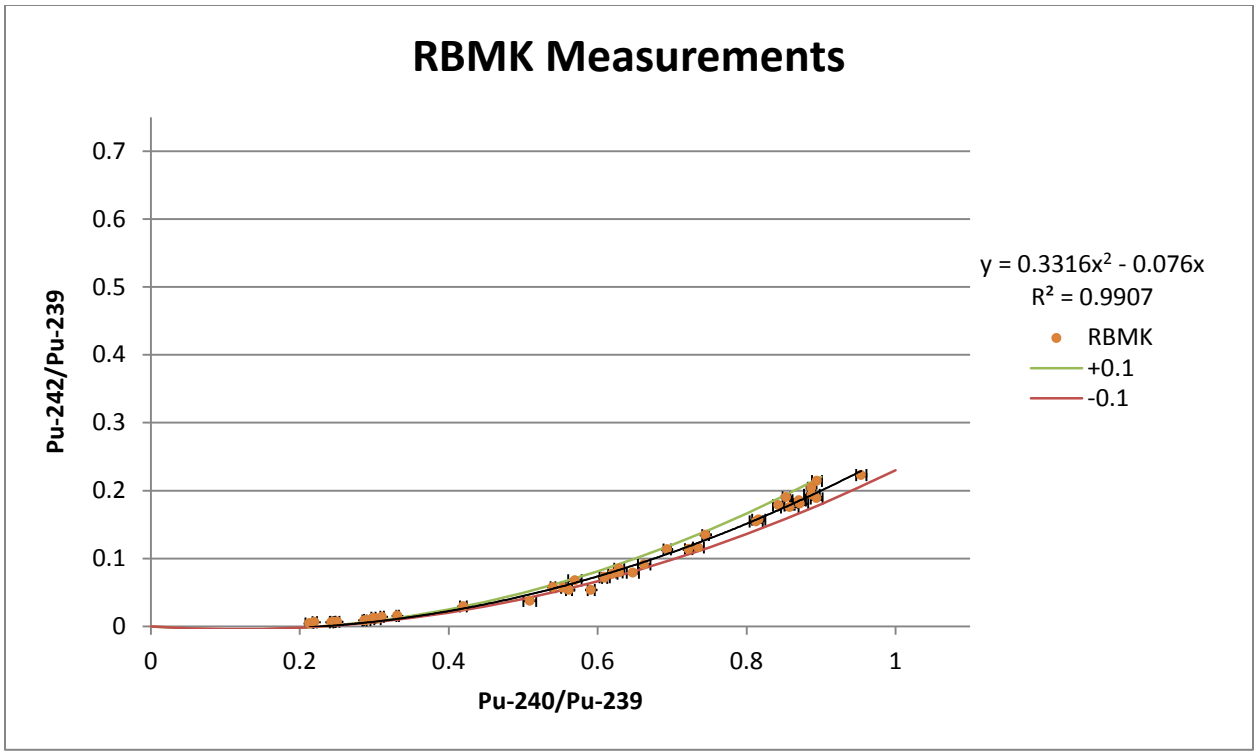

Figure 25 RBMK Pu-242/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +10% and -10% Uncertainty Bands

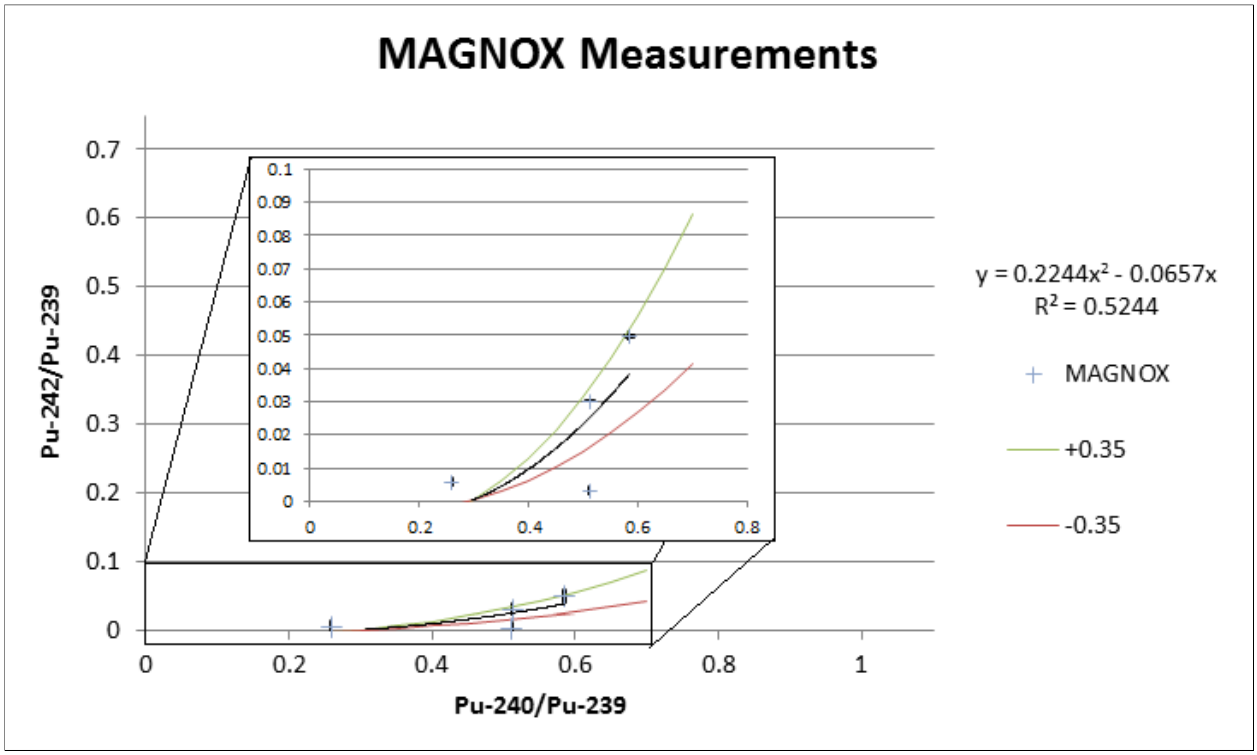

Figure 26 MAGNOX Pu-242/Pu-239 Measurement Data with Error Bars and a Second Order Least Squares Fit with a +35% and -35% Uncertainty Bands

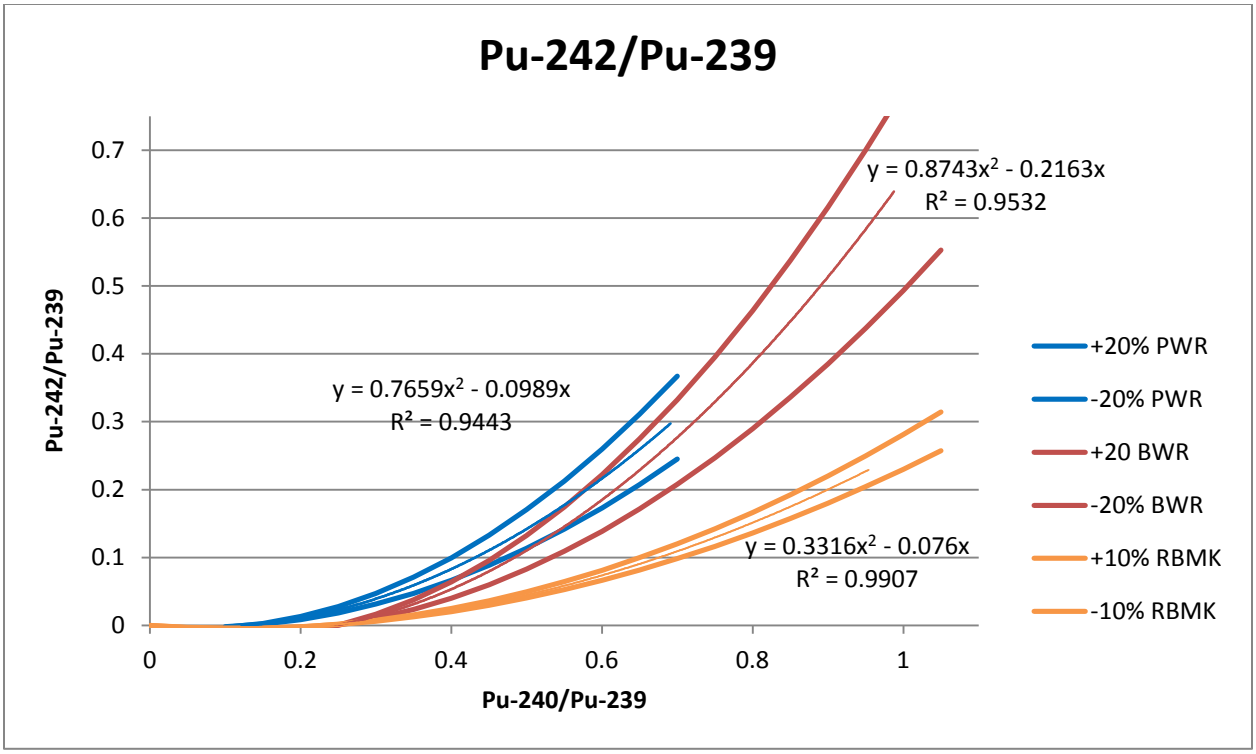

Figure 27 PWR, BWR, and RBMK Pu-242/Pu-239 Measurement Data Comparison with Second Order Least Squares Fits and Uncertainty Bands

One thing to notice is that while the graphite PRs were operated at a much lower burnup than the BWR and PWR reactors, their ratios match up well with the literature. The graphite PRs operated at much lower burnups because the mission of the reactors was to produce plutonium with a Pu-240/Pu-239 ratio of 0.06 or less. However, in some instances, higher burnups were also achieved in these reactors for the purpose of understanding the physics of the reactors and can be seen in the dataset. There is a noticeable difference in data range for the PRs and the light water reactors. PRs were operated to a much lower burnup and the majority of the graphite PR data has a Pu-240/Pu-239 ratio of less than 0.2. There was also no Pu-238 or Pu-240 information for some of the PWR reactors and no Pu-238 information for the CANDU reactors and some of the BWR reactors. The data range in which there was no information corresponded to lower burnup fuel which may have put the Pu-238 below the detection limit of the measurement technique at that time.

SFCOMPO data contains a large number of measured parameters; however measured data for all of the isotopes of interest are not included for every reactor type. In order to obtain data for these isotopes, calculations were performed and compared to the measured data to determine if the models accurately represented the measured data. Table 4 shows a comparison of the Leningrad RBMK-1000 reactor assembly R-1029. The measured data column contains all of the measurements completed by the laboratory in Khlopin, the calculated column gives the calculational data generated by the ORIGEN-ARP[12] computer code, and the final column gives the ratio of measured compared to calculated data. This

[12] ORIGEN-ARP: Automatic Rapid Processing for Spent Fuel Depletion, Decay, and Source Term Analysis. Oak Ridge National Laboratory. ORNL/TM-2005/39 Version 6.1. June 2011.

sample represents fuel that was initially enriched to 1.8%, burned to a burn-up of 16.8 GWD/MTU, and cooled for 4.6 years prior to measurements being taken.

Table 4. Comparison of Measured and Calculated Data of the Leningrad RBMK-1000

Reactor Type	Leningrad		RBMK-1000		
Assembly			R-1029		
Initial U-235 Enrichment			1.8	1.8	
Cooling Time (years)			4.6	4.6	
Measurement Lab/Code			Khlopin	ORIGEN-ARP	
	Units		**Measured**	**Calculated**	**E/C**
Am-241	kg/MTU initial		0.00025	0.1225	0.002041
Am-243	kg/MTU initial		0.15	0.02022	7.418398
Burnup			16.79733718	16.8	0.999841
Cm-242	kg/MTU initial		0.065	0.000003756	17305.64
Cm-244	kg/MTU initial		0.0298	0.00206	14.46602
Np-237	kg/MTU initial		1.03	0.1	9.894332
Pu-238	kg/MTU initial		0.0292	0.03	1.093633
Pu-239	kg/MTU initial		2.615	2.43	1.074805
Pu-240	kg/MTU initial		1.644	1.52	1.080158
Pu-241	kg/MTU initial		0.511	0.44	1.170408
Pu-242	kg/MTU initial		0.222	0.23	0.98579
U-234	kg/MTU initial		0.0886	0.12	0.760515
U-235	kg/MTU initial		4.865	4.9	0.99225
U-236 Build up	kg/MTU initial		2.07	2.15	0.963239
U-238	kg/MTU initial		969.8	970.4	0.999382

The uranium and plutonium production shows fairly good agreement (the E/C ratio is close to 1), however the other minor actinides do not agree. Table 4 represents one measurement on one specific assembly. In order to determine if this trend exists for other assemblies and/or reactors types, it would require many more calculations and are outside of the scope of this current report and could be investigated later. This comparison using plutonium isotopics indicates that PRs can be identified from other reactor types and basic operating parameters may be determined, as isotopic ratios of the plutonium vector vary with different moderator temperatures. However, there may be better comparisons using other isotopes such as minor actinides or fission products that will allow for the further identification of the reactor types and operating history.

8.0 Measurement Techniques

A method for the identification of observable radionuclides from neutron activation analysis (NAA) of dissolved spent nuclear fuel samples was also investigated.[13] Spent fuel samples used in the NAA had variable burnup values and cooling times on the order of 20 - 30 years. They were first modeled using ORIGEN-ARP to estimate nuclide concentrations. Concentrations were used as input for activation calculations to determine the viability of nuclide detection given assumed experimental parameters. Spent fuel parameters that were modeled such as burnup and cooling time were chosen to be representative of spent fuel samples provided by PNNL to the University of Texas at Austin. The model was subsequently tested by performing NAA on the spent fuel samples at the Nuclear Engineering Teaching Laboratory (NETL) at the University of Texas at Austin.[14] Results indicate NAA was successful in identifying several stable and long-lived radionuclides predicted via model calculations but results appear limited to sample concentration.

The predictive model was formulated and nuclear decay data were used to predict neutron activation analysis results of the two dissolved spent nuclear fuel samples provided by PNNL with variable burnup values and cooling times. Model predictions were tested by performing thermal instrumental neutron activation analysis on the spent nuclear fuel samples using both cyclic and conventional irradiation methods.

NAA can be used to detect stable and long-lived nuclides (whose very slow to nonexistent decay rates provide difficulty for passive spectroscopy techniques) by activating these problematic nuclides with higher specific activity and signal output. NAA has the capability to quantify multiple nuclides and elements in a single experiment run and is also non-destructive and a relatively rapid analysis technique. It is expected that accurate analysis of the noble metal phase will also tell what kind of fuel it came from and how it was chemically processed. PNNL had several samples of noble metal phase separated from commercial fuel using two different reprocessing schemes. This phase may include fission products such as silver, cadmium, selenium, and tellurium, in addition to the noble metals. Only a few papers have been published on chemical analysis of the noble metal phase. If the noble metal phase tends to collect tellurium, for example, then the ratio of rhodium or palladium to tellurium might provide a good indication of the amount of plutonium in the fuel.

Neutron activation analysis was performed on two dissolved samples extracted from the high burnup, commercial UO_2 fuel Approved Testing Material (ATM-109) for identification of the stable and quasi-stable noble metal nuclides. ATM 109 was produced using fuel irradiated in reactor I at the Quad Cities nuclear power plant (NPP). The fuel was fabricated by General Electric for use in 7×7 assemblies. Fuel was 3.0 % enriched and irradiated between February 1979 and September 1987 and again between November 1989 and September 1992 with an approximate burnup of 67 MWd/kgU - 70 MWd/kgU. As of early 2013, fuel has been cooling for approximately 21 years.

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[13] *Predictive Modeling of Neutron Activation Analysis of Spent Nuclear Fuel for the Detection of Stable and Long-Lived Radionuclides*. Palomares, RI, et al. (2012), University of Texas at Austin.

[14] *Measuring the Isotopic Composition of Extracted Noble Metal Phase from Used Nuclear Fuel*. Palomares, RI, et al. (2013), University of Texas at Austin.

FY12 activities included measurement of separated long-lived and stable fission product elements, starting with the noble metal phase; which are an alloy of metallic molybdenum, technetium, ruthenium, rhodium, and palladium (Mo, Tc, Ru, Rh, and Pd). Because the noble metal phase is chemically unreactive and remains undissolved after the fuel has been reprocessed, it may be able to provide an indicator that spent fuel was once present.

Two different processing methods had been applied to derive the PNNL samples. The first one was the fuel was dissolved by carbonate-peroxide leaching process and the one, the fuel was dissolved by hot nitric acid leaching. Both sets of resulting un-dissolved solids were examined (See Figure 28). These two scanning electron microscope (SEM) pictures show the noble metal phase and that the epsilon metal particles are exceptionally small. The nature of the particles was revealed by observing them at low energy and with secondary electron imaging.

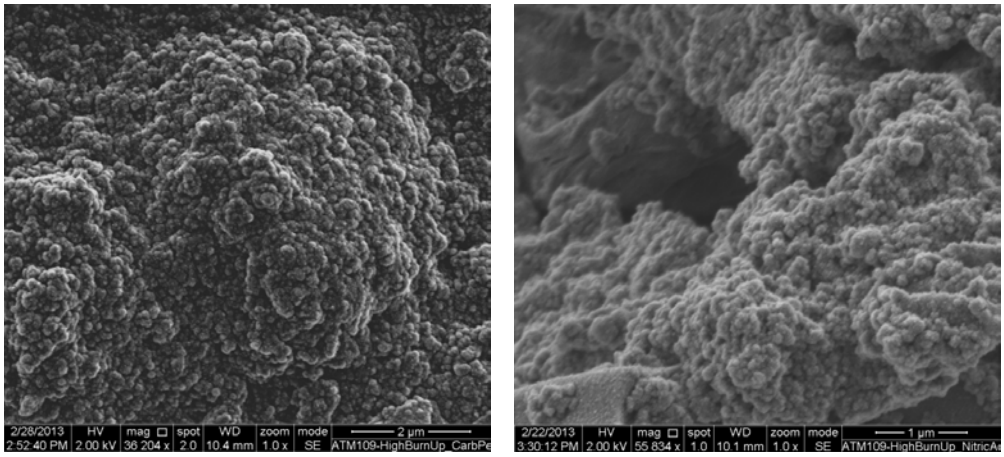

Figure 28. A. Carbonate-Peroxide, Sample C B. Hot Nitric Acid, Sample N

Particles up to a few microns in diameter were recovered from used fuel, slurried in water, and placed in polyethylene vials. Slurries were then dried into opaque crusts at the bottom of the vials. Samples differ only by chemical solvent used to dissolve the UO_2. The first sample (sample C) was prepared from used fuel dissolved in an ammonium carbonate and hydrogen peroxide solution whereas the second sample (sample N) was prepared from used fuel dissolved in nitric acid. Actinides and other fission products in solution were chemically separated from the undissolved noble metal phase but quantities of fission products and transuranic elements remain. Sample C weighs approximately 0.8 mg and sample N weighs approximately 2.0 mg.

Sample N was examined to estimate intrusive fission product quantities. Total plutonium content was estimated at approximately a microgram. Total uranium content was estimated at no more than ten micrograms and ^{235}U content was estimated to be very low due to very high fuel burnup. Noble metal phase was estimated to be ten to twenty percent ^{99}Tc (~7µg in sample N). ^{90}Sr was reported to be in approximately equal ratio to ^{137}Cs. The sample was also counted for measurement of gamma emitters.

Mass quantities of ^{98}Mo, ^{100}Mo, ^{102}Ru, ^{104}Ru and ^{103}Ru were obtained with an uncertainty level of two-sigma. Sources of uncertainty include counts, efficiency, gamma ray intensity, decay constant, cross section, neutron flux, irradiation time and decay time.

Comparative peak analysis was successful in identifying 5 of the 8 noble metals considered to be viable analytes for NAA. Peaks corresponding to ^{99}Mo, ^{101}Mo, ^{103}Ru, ^{105}Ru and ^{104m}Rh were observed. Peaks corresponding to ^{104}Rh and ^{109}Pd were also observed, but eliminated from further consideration due to poor counting statistics and/or interferences with intrusive fission product lines; ^{104}Rh was rejected due to interference with ^{91}Sr at 555 keV and ^{109}Pd was rejected due to interference with ^{155}Eu at 188 keV. Uncertainties are relative to specific spectra, nuclide, and gamma ray energy; therefore, uncertainty ratios for the highest intensity gamma rays for all nuclides were collected and averaged. From the results of this study, it shows the errors associated with the experiment (counts, decay and irradiation time, and efficiency factors) were relatively low in comparison to the nuclear data, which indicates good experimental data.

In addition, Instrumental NAA has the potential for high sample-processing rates and rapid analysis. Separation of the noble metal phase from the actinides and remaining fission products greatly improved NAA results in comparison to preliminary results from NAA of diluted ATM, performed in earlier in 2012. Having implemented an experimental protocol, numerous noble metal phase samples (similar to the ones studied here) may be analyzed in a matter of hours.

9.0 Next Steps

Follow on work for FY14 will focus on identifying the gaps of information in the dataset and working to fill them as well as developing methods to identify reactor types of origin. As new measured data becomes available, it will continue to be incorporated into the dataset. However it is assumed unlikely that any significant quantity of new measured data will be identified for incorporation since all the open-source data identified in the FY10 literature search (and more) have now been incorporated. The primary effort in FY14 will be to perform additional calculations to supplement the measurements and start more detailed data analyses, such as identifying if other isotopes including actinides and fission products allow for better identification of reactor type. The calculated data will address concerns that the measured data for each reactor type may not adequately address the range of credible operating conditions for that reactor type. The priority of reactor types has been selected based on the number of units of each reactor type that have been in operation worldwide. Table 5 was used for the selection process, and shows the distribution and variety of reactor systems as of 2011. A brief description of each of these reactor systems can be found in Appendix B.

The SFCOMPO/NF dataset is limited in that the inventory data (i.e., isotopic ratios) do not fully cover the anticipated range of operating conditions for these reactors. Since operating conditions, enrichments, etc. can vary from one country to another, it is necessary to have a wider range of data to successfully apply comparisons for nuclear forensics applications. The range of burnups and enrichments in the measured SFCOMPO/NF data are not inclusive enough to consider them "representative" of all fuels. This representativeness is a critical parameter for establishing the degree of confidence in "matching" a specific sample to a reactor type. Essentially, one can think of the most common operating conditions as representing the most probable match to a reactor type. Calculating the impact or sensitivity of an isotopic ratio across the range of credible operating conditions from the norm to extreme provides an estimate of the decrease in confidence of a "match". The uncertainty in the measured data itself and the comparison of the measured to the calculated data also impact this confidence. Increasing the quantity of isotope ratio data to include a realistic range of operating conditions is necessary to provide a basis for the use of these data for nuclear forensics purposes. The degree of confidence or the probability of a match between an interdicted sample and a given reactor type/source must be quantified. Figure 4, Figure 12, and Figure 20 demonstrate that there is overlap in data from different reactor types. This overlap is expected because of the large range of operating conditions and can be assessed by either requiring multiple isotopic ratios to provide consistent indication of a match to a specific reactor type and/or require a statistical analysis to establish the confidence/quality of the attribution.

The focus of the FY14 activities will be to develop calculated data to represent the range operating conditions and incorporate available uncertainty data. This extension of the SFCOMPO/NF dataset is a necessary step in preparation to establish an algorithm for quantitatively establishing attribution of an interdicted sample to a specific reactor type. Using the dataset, specific "reactor-type" models will be developed with isotope ratios of interest. These models will be incorporated into a classification algorithm and using multi-dimensional regression analysis to identify reactor types of origin.

Table 5. Power Production Reactors Worldwide
(bold numbers are currently operating reactors as of 2011;
italic numbers are permanently shut down or decommissioned)

Country	AGR	BWR	FBR	Gas Graphite	GCHWR	HTGR	LWCHWR	LWGR	LWGR (RBMK)	MAGNOX	PHWR	PHWR (CANDU)	Production	PWR	PWR (VVER)	Sodium graphite	SGHWR
Argentina											**1**	**1**					
Armenia															**1**,*1*		
Belgium														**7**,*1*			
Brazil														**2**			
Bulgaria															**2**,*4*		
Canada												**18**,*5*					
China						**1**						**2**		**7**	**2**		
Czech Republic															**6**		
Finland		**2**													**2**		
France			**1**,*1*	*9*	*1*									**59**,*1*			
Germany		**6**,*5*	*1*		*1*	*2*					*1*			**11**,*4*	*5*		
Hungary											**15**				**4**		
India		**2**															
Italy		*2*								*1*				*1*			
Japan		**32**,*1*	**1**				*1*			*1*				**23**			
Kazakhstan			*1*														
Korea (ROK)											**4**			**16**			
Lithuania									**1**,*1*								
Mexico		**2**															
Netherlands		*1*												**1**			
Pakistan												**1**		**1**			
Romania												**2**					
Russia		*1*	**2**						**11**,*3*				*14*		**15**,*2*		
Slovakia					*1*										**5**,*1*		
Slovenia														**1**			
South Africa														**2**			
Spain		**2**		*1*										**6**,*1*			
Sweden		**7**,*2*												**3**			
Switzerland		**2**			*1*									**3**			
Taiwan		**4**												**2**			
United Kingdom	**14**,*1*		*2*			*1*				**4**,*22*				**1**			*1*
Ukraine									*4*						**15**		
United States		**35**,*7*	*1*			*1*		*1*			*1*		*14*	**69**,*9*		*1*	
TOTAL	**15**	**113**	**10**	**10**	**4**	**5**	**1**	**1**	**20**	**28**	**21**	**29**	**28**	**231**	**65**	**1**	**1**

10.0 Conclusions

A baseline version of SFCOMPO/NF has been established using all identified open-source data. Additions to the OECD/NEA SFCOMPO data included measured data found in the open literature and historical Graphite Hanford production reactor records, as well as calculated values from models. Initial comparisons of plutonium isotopic ratios clearly demonstrate the potential of SFCOMPO/NF to be an important tool for nuclear forensics. Investigations were also performed to evaluate NAA as a potential experimental method that could provide additional measurements to enhance SFCOMPO/NF and/or to provide for the rapid analysis of interdicted samples.

FY13 work contributed further analysis to the expanded dataset from FY12 along with the addition of the available measurement uncertainties which was able to put data spread and fit confidence in context by showing that the data spread is much larger than data uncertainty.

Neutron Activation Analysis appears a viable analysis technique for the detection of stable and long lived nuclides such as ^{238}U, ^{99}Tc, and ^{109}Ag in spent nuclear fuel. These nuclides and other fission products could potentially be exploited as additional markers for establishing a match between interdicted samples and a given reactor type or may provide insight into additional characteristics of sample such as age, processing method or country of origin. The NAA results to date are limited by sample mass and activity. To further validate neutron activation as an effective analysis technique for spent nuclear fuel and interdicted samples, spent fuel samples of higher concentration would be needed for the experiment. Such samples would have higher activities than the samples used to date which would also permit us to gauge the limits of NAA as a spent fuel analysis technique.

Uncertainty estimates for both calculations and experiments are necessary components for evaluating the probability/significance of a match between interdicted material and a specific reactor type. Simple models using tools such as ORIGEN-ARP will be most efficient to examine the impact of the range of operating conditions on the mass ratios for different reactor types. Quantifying variations in the isotopic ratios that occur within a specific reactor type due to operations and core location will require a more detailed calculations using a lattice physics code(e.g., WIMS or TRITON). Emphasis for calculating any additional data will be placed on reactor samples that will give the most complete representation of operating conditions.

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Appendix A

Reactor Measurements Added to SFCOMPO/NF Dataset

Appendix A

Reactor Measurements Added to SFCOMPO/NF Dataset

Appendix A summarizes reactor samples that were added which are not currently in the SFCOMPO dataset.

A.1 Pressurized Water Reactors

A.1.1 Three Mile Island

Three Mile Island, a PWR located in Pennsylvania, has two separate units; the isotopic composition data is concerned with Unit 1. Wolf et al. (2008) presents isotopic compositions of uranium, plutonium and americium along with several lanthanides measured by inductively coupled plasma-mass spectrometry (ICPMS), γ-spectrometry (γ-S), and α-spectrometry (α-S) from one fuel rod (five samples in all). These measurements are for burnups ranging from 44.8 – 51.3 GWd/MTU with an initial ^{235}U enrichment of 4.0 wt%. A second set of five samples from the same fuel rod (burnup range: 44.8 – 55.7 GWd/MTU) is presented in Wolf et al. (2000), with extensive operating history available in a calculation package generated by the Office of Civilian Radioactive Waste Management (Scaglione (2002)). In a third source (TRW Yucca Mountain Project Test Report (1999)), eight more samples from three fuel rods were analyzed for uranium, plutonium, americium and several lanthanides by isotope- dilution mass spectrometry (IDMS). The samples had burnups from 22.8 – 29.9 GWd/MTU at an initial ^{235}U enrichment of 4.67 wt%. No specific operating history is given in this document.

A.1.2 Turkey Point

Turkey Point nuclear generating station is a two-unit PWR located in Florida. Davis and Pasupathi (1981) report a detailed operating history for Turkey Point fuel rods G7, G9, J8, I9, and H6. The concentration of ^{148}Nd and the isotopic compositions of uranium and plutonium are determined by mass spectrometry. The fresh fuel had an enrichment of 2.559 wt% ^{235}U and burnup values ranging from 19 to 28 GWd/MTU. Atkin (1981) performs similar analyses on fuel rods G9, G10, and H9.

A.1.3 Gösgen

Gösgen nuclear power plant (in German: Kernkraftwerk Gösgen, or KKG) is a PWR built near Däniken, Switzerland by the German company Kraftwerk Union AG, a former subsidiary of Siemens that is now part of AREVA NP. The isotopic compositions of samples from Gösgen were also measured in the ARIANE program. These measurements are the same as measured for Dodewaard and Beznau samples and are described in Primm (2002). Four samples from two different assemblies were examined. The first two samples had an initial enrichment of 3.4 wt% 235U (each samples from a different fuel rod) and burnups of 32.5 and 59.7 GWd/MTU. The other two samples were from the same fuel rod, with 4.1 wt% ^{235}U and burnups of 29.1 and 52.5 GWd/MTU.

Grambow et al. (1997) examine a high burnup fuel pellet from Gösgen using traditional radiochemical separations and analysis techniques. Three different sections of the pellet are analyzed: the center region, the outer edge, and the remainders left on the cladding surface after pellet extraction. Samples from each region are approximately one millimeter in diameter and dissolved samples were analyzed by two independent laboratories. Increased fission product concentrations in the fuel remainders confirm the presence of a high burnup rim. Basic details regarding burnup, average linear power, irradiation time, etc. are provided

A.2 Boiling Water Reactors

A.2.1 Quad Cities

Quad Cities is a BWR located in Illinois that also has two units. Wolf et al. (2008) repeats the measurements performed for Three Mile Island, Unit 1 for seven samples from Quad Cities, Unit 1. The seven samples are spread across three fuel rods:

- three samples from a rod with a mean burnup of ~70 GWd/MTU and 3.0 wt% initial ^{235}U UO_2;
- two samples from a rod with a mean burnup of ~62 GWd/MTU and 3.8 wt% initial ^{235}U UO_2; and
- two samples with a mean burnup of 65 GWd/MTU and 2 wt% Gd2O3-3.0 wt% ^{235}U UO_2.

There is some question regarding the validity of this data, however, as the mass balances did not close. It is suspected that the sample preparation method added significant amounts of iron to the sample and thus skewed the results. As with the Three Mile Island data, six more samples were analyzed and presented in the TRW Yucca Mountain Project Test Report (1999). These samples were distributed as follows:

- one sample from a rod with a burnup of 55.0 GWd/MTU and 3.0 wt% initial ^{235}U UO_2;
- four samples from a rod with local burnups of 52.5, 53.1, 70.0 and 74.6 GWd/MTU and 3.8 wt% initial ^{235}U UO_2; and
- one sample with a burnup of 45.5 GWd/MTU and 2 wt% Gd2O3-3.0 wt% ^{235}U UO_2.

Though not stated as such, these sample sets appear to be from the same fuel rods. To date, no operating history information has been found for these particular samples. A third set of isotopic composition data is provided in Fisher and Difilippo (1998). This dataset contains measurements of uranium, plutonium, neptunium, americium, curium and neodymium by an unspecified method for 9 fuel rods of various types (4 of which have mixed oxide fuels). The burnups ranged from 7.2 to 13.4 GWd/MTU. An extensive operating history is available in a supplementary document.

A.3 Production Reactors

A.3.1 B Reactor

The B Reactor at the Hanford Site was the first large-scale nuclear reactor ever built. The project was commissioned to produce plutonium-239 by neutron activation as part of the Manhattan Project. B reactor was a light water cooled, graphite moderated reactor fueled with metallic uranium.

The power levels of B Reactor increased through the years of operation starting at 250 MWth and ending up at ~2000 MWth. This increase in power has an effect on the production of plutonium and other higher actinides. B Reactor was originally fueled with natural uranium, however to increase power levels and maintain the mission of plutonium production, enriched and recycled uranium was used which also has an effect on the isotopics of the spent fuel.

A.3.2 K East and K West Reactors

Hanford's "sister reactors", the K-East and the K-West Reactors, were built side-by-side in the early 1950's. The two reactors went operational within four months of each other as K-West went into service in January of 1955 and K-East started operations in April of that same year. K-West was the seventh reactor built at Hanford; K-East was the eighth. Like B Reactor, the primary goal of these two reactors was plutonium production. These reactors operated at higher powers and utilized natural, enriched, and recycled uranium fuel.

A.3.3 C Reactor

Hanford's C Reactor was built in the late 1940's and early 1950's, and started operations in 1952. It was the sixth reactor built at Hanford, and ran for sixteen years before being shut down in 1969. C Reactor was designed to operate at higher power levels than the previous reactors built. Experimental fuels and targets were used in C Reactor in addition to the natural, enriched, and recycled uranium.

A.3.4 Heavy Water Production Reactors from Savannah River Site.

All five reactors built at Savannah River Site (SRS) were heavy water moderated designs, and they could be reconfigured for a variety of missions. The most important mission for these reactors was the production of both plutonium and tritium for the weapons program. In addition, they were able to produce Cf-252 for neutron sources, Cm-244 for heat sources for RTGs and Pu-238 also for RTGs. R reactor was the first to start-up in December of 1953, and it shutdown in June of 1964. P, L and K reactors all started in 1954 (February, July and October respectively). L reactor was shut down in June of 1988, followed by P reactor shutting down in August of 1988. K reactor didn't shutdown until July of 1992. C reactor was the last to start-up in March of 1955, and it shutdown in June of 1985. Typically, these reactors used low enriched uranium metal for fuel.

A.4 CANDU Reactors

A.4.1 NPD CANDU Reactor

The Canadian Nuclear Power Demonstration (NPC) Reactor at Rolphton, Ontario was the first small scale CANDU prototype (22MWe) that went into operation in 1962. NPD reactor fuel consists of a 19 element bundle consisting of a central fuel pin, surrounded by 6 middle and 12 outer fuel pins, respectively. The fuel material is natural uranium with Zircaloy cladding.

A.4.2 Bruce CANDU Reactor

The Bruce-A Nuclear Generating Station Reactor is a 750 MWe heavy water reactor that first went operational in 1977. Bruce reactor fuel bundles have 37 fuel pins, consisting of a central fuel pin surrounded by three concentric rings of 6, 12 and 18 fuel pins, respectively. The fuel material is natural uranium with Zircaloy cladding.

A.4.3 Pickering CANDU Reactor

The Pickering-A Canadian Nuclear Power Reactor, a 515MWe heavy water reactor, was Canada's first commercial CANDU reactor that began operations in 1971. Pickering reactor fuel consists of a 28 fuel pins, consisting of 4 inner fuel pins, surrounded by 8 middle and 16 outer fuel pins. The fuel material consists of natural uranium with Zircaloy cladding.

A.5 MAGNOX Reactors

A.5.1 Hunterston Reactor

The Hunterston MAGNOX reactor is 160 MWe plant in the United Kingdom that started up in 1964 and shutdown in 1990. It is a graphite-moderated, natural uranium fueled reactor that is cooled with CO_2. Uranium fuel elements are inserted into vertical channels in this graphite core. The uranium fuel is clad in a special magnesium alloy hence the name 'Magnox'.

A.5.2 Bradwell Reactor

The Bradwell MAGNOX reactor is 123 MWe plant in the United Kingdom that started up in 1962 and shutdown in 2002. It is a graphite-moderated, natural uranium fueled reactor that is cooled with CO_2. Uranium fuel elements are inserted into vertical channels in this graphite core. The uranium fuel is clad in a special magnesium alloy hence the name 'Magnox'.

A.6 VVER Reactors

A.6.1 Novovoronezh NPP-3 Reactor

A Pressurized Water Reactor of Russian design (***V***odo-***V***odyanoi ***E***nergetichesky ***R***eaktor, or “Water-water energetic reactor,” i.e. water cooled and water moderated). Novovoronezh NPP-3 is a VVER-440 that began commercial operation in 1972.

A.6.2 Novovoronezh NPP-4 Reactor

A Pressurized Water Reactor of Russian design (***V***odo-***V***odyanoi ***E***nergetichesky ***R***eaktor, or “Water-water energetic reactor,” i.e. water cooled and water moderated). Novovoronezh NPP-4 is a VVER-440 that began commercial operation in 1973.

A.6.3 Novovoronezh NPP-5 Reactor

A Pressurized Water Reactor of Russian design (***V***odo-***V***odyanoi ***E***nergetichesky ***R***eaktor, or “Water-water energetic reactor,” i.e. water cooled and water moderated). Novovoronezh NPP-4 is a VVER-1000 that began commercial operation in 1981.

A.6.4 Kalinin Reactor

A Pressurized Water Reactor of Russian design (***V***odo-***V***odyanoi ***E***nergetichesky ***R***eaktor, or “Water-water energetic reactor,” i.e. water cooled and water moderated). Kalinin is a VVER-1000 that began commercial operation in 1985.

A.6.5 Balakovo Reactor

A Pressurized Water Reactor of Russian design (***V***odo-***V***odyanoi ***E***nergetichesky ***R***eaktor, or “Water-water energetic reactor,” i.e. water cooled and water moderated). Balakovo is a VVER-1000 that began commercial operation in 1986.

A.7 RBMK Reactors

A.7.1 Leningrad Reactor

Leningrad 1 is a 925 MWe *Reaktor Bolshoy Moshchnosti Kanalniy*, "High Power Channel-type” (RBMK) Reactor. It began operation in 1973 and went into commercial operation in 1974. It has been off-line since May 2012 because of deformation of its graphite moderator. Fuel is slightly enriched uranium oxide with a Zircaloy tube around it.

Appendix B

Description of Power Reactor Types

Appendix B

Description of Power Reactor Types

Below is a description of the power reactor types listed in Section 9.0, Table 5.

AGR: **A**dvanced **G**as-cooled **R**eactors (Figure B.1) are the second-generation graphite-moderated, gas-cooled reactors built in Great Britain. AGRs use slightly enriched (< 4%) UO_2 pellets in stainless steel cladding. CO_2(g) is the primary coolant and water removes heat from 650°C CO_2 to turn steam turbines. (Nuclear Power Reactors 2010)

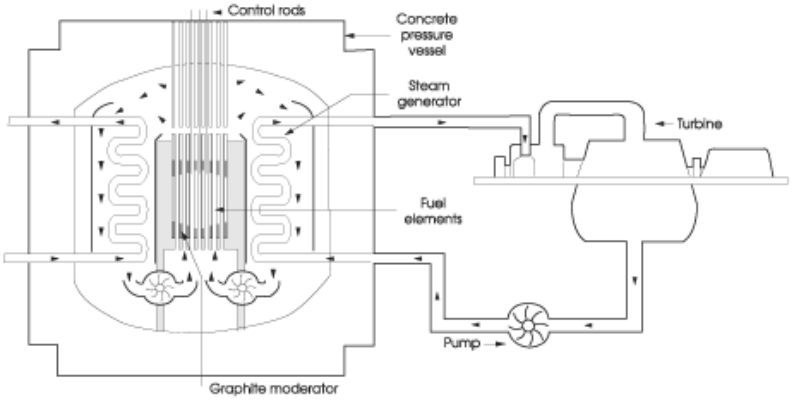

Figure B.1 Advanced Gas-Cooled Reactor (Nuclear Power Reactors 2010)

BWR: Pioneered in the United States by General Electric, the boiling water reactor allows the heated, light water coolant/moderator to become steam in the reactor vessel (Figure B.2, right). The ~22-meter tall vessel contains control rods and drives below the reactor core and steam driers above the core that allow direct transfer of steam to the turbines. BWR fuel assemblies generally contain 8 x 8 or 10 x 10 fuel pins and are positioned adjacent to cruciform-shaped control rods. Fuel rods at assembly edges have lower ^{235}U enrichment than those in the center of the assembly and several fuel rods in the center of the assembly contain water instead of fuel in order to flatten the power distribution. Each fuel rod is made of zircalloy cladding and each assembly has a zircalloy fuel channel to prevent large void areas in the core. Reactivity is controlled in the short term by recirculation flow rates and poison control rods and in the long term by poison control rods and burnable poisons in the fuel pellets (e.g., Gd_2O_3).

FBR: Fast breeder reactors use a fast neutron spectrum to fission ^{235}U for heat generation and ^{238}U, either in a surrounding blanket or in the seed fuel matrix, to produce fissile ^{239}Pu. The use of fast neutrons prohibits the use of water as a coolant because of its substantial moderating properties. Fast breeder reactors have used very efficient liquid metal coolants such as sodium, sodium-potassium, and lead-bismuth.

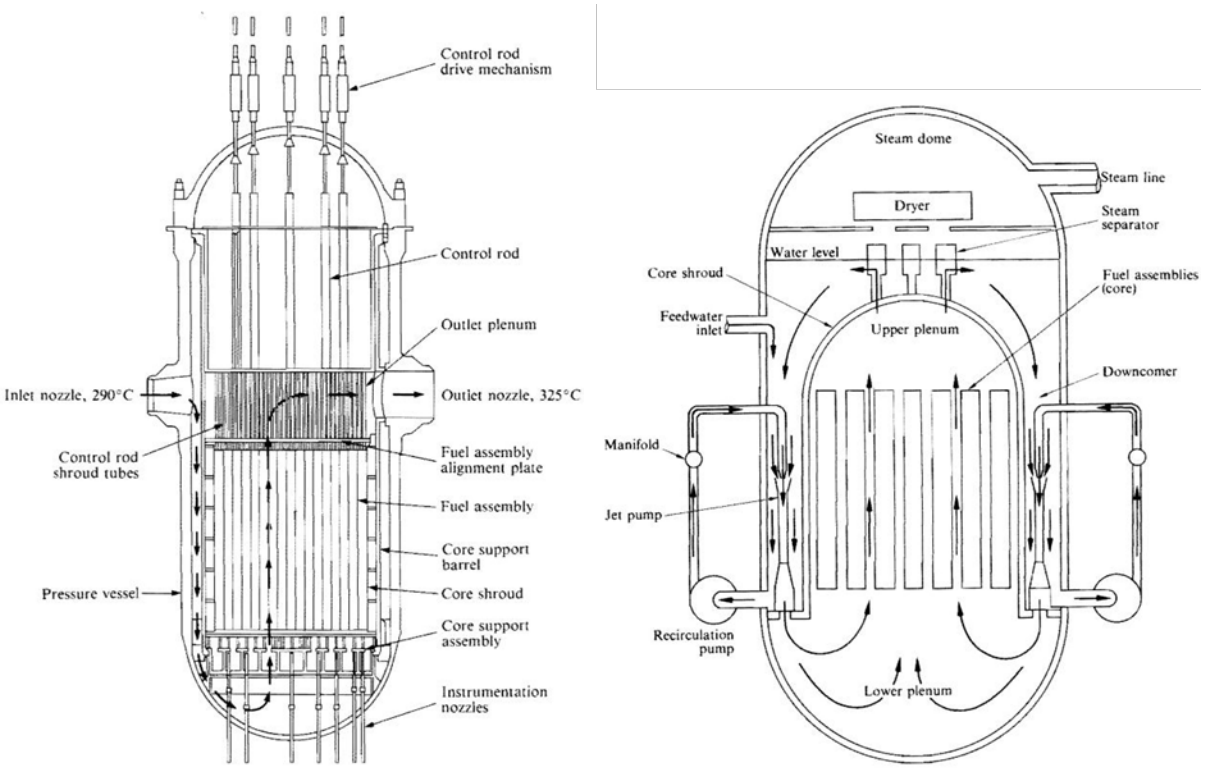

Figure.B.2 Generalized Diagrams of a PWR (left) and a BWR (right) (Lamarsh 1983C)

Gas graphite: Reactors that are gas cooled and graphite moderated. See entries for AGR and MAGNOX.

GCHWR: Gas cooled heavy water reactors use a gas (e.g., He, CO_2) to remove heat and heavy water to moderate neutrons.

HTGR: A high temperature gas-cooled reactor uses oxide or carbide fuel microspheres coated in various moderating or structural materials. Cooled by helium gas, the core can be configured in two ways: the microspheres are embedded in a graphite pebble which is used in a hopper- like core configuration, or the microspheres are compressed with a binder into long rods for use in a core moderated with graphite blocks.

LWCHWR: Light water cooled heavy water reactors are cooled with H_2O and moderated with D_2O. An SGHWR is an example of a LWCHWR.

LWGR: Light Water Graphite Reactors are light water cooled and graphite moderated.

LWGR (RBMK): A Russian Light-Water Graphite Reactor (***R***eaktory ***B***olshoy ***M***oschnosti ***K***analniy, or "High power channel-type reactor") using UO_2 fuel with 1.8 to 2% enrichment. Light-water coolant flows in pressure tubes through moderating graphite blocks. The RBMK design allows the water coolant to boil, causing void spaces and a positive reactivity coefficient. The RBMK's large size and fuel loading can cause difficult reactivity control due to multiple, loosely coupled critical masses in the core. The reactors at Chernobyl were RBMK reactors.

LWR: Light water reactor is a general term for nuclear power reactors using H_2O as a coolant and moderator, including BWRs and PWRs.

MAGNOX: A CO_2(g)-cooled, graphite moderated reactor design from the United Kingdom. Natural uranium metal rods are housed in "cans" made of a ***mag***nesium ***n***on-***ox***idizing alloy. About a meter long, multiple magnox cans with different heat transfer fins were developed over the lifetime of the program.

PHWR: Pressurized heavy water reactors use deuterium as both moderator and coolant. The heavy water may be managed using one large pressure vessel or with many (~102) pressure tubes.

PHWR (CANDU): ***Can***ada ***D***euterium ***U***ranium (CANDU) reactors comprise the bulk of PHWRs. CANDU reactors use pressure tubes to separate moderator and coolant volumes. As shown in Figure B.3, CANDU reactors use a calandrial reactor vessel (7.6-m diameter and 4-m depth) with 380 pressure tubes containing 12 natural uranium, zirconium-clad fuel bundles placed sequentially down the length of the tubes. Two separate water coolant systems transport D_2O at ~310°C through the pressure tubes and the fuel bundles. Each coolant loop moves half of the total D_2O coolant through adjacent pressure tubes in opposite directions. A refueling machine allows addition and removal of fuel bundles in one pressure tube at a time without halting coolant flow or requiring reactor shutdown. This ability to refuel on-line provides finer reactivity control because it does not require 18 to 24 months of reactivity to be added all at the same time, as happens in LWRs. A separate moderator system moves heavy water past the outside of the pressure tubes at a low temperature (~70°C) to reduce the loss of costly D_2O that occurs in the higher temperature coolant loops.

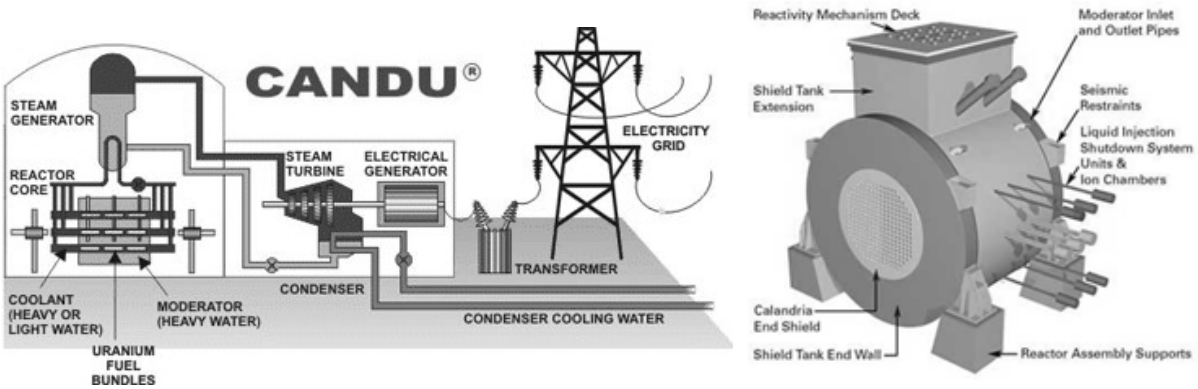

Figure B.3 A general schematic of a CANDU reactor (left) (CANDU Technology) and the calandria (right) (CANDU Calandria)

Production: A reactor used for producing plutonium.

PWR: Pressurized water reactors were originally developed in the United States by Westinghouse (Figure B.2). PWRs have a closed primary loop, where a ~13-meter high reactor vessel is pressurized to ~15.5 MPa to prevent the water coolant/moderator from boiling. Heat is transferred to one or more steam loop(s) via heat exchangers, allowing the secondary loop(s) to be free from reactor core contamination. Newer PWR fuel assemblies are typically 17 x 17 arrays of zircalloy fuel pins containing slightly enriched UO_2 fuel pellets. Short term reactivity is controlled by full-length and part-length (or axial power

shaping) rods positioned above the core. Intermediate and long term reactivity is controlled by a soluble poison (e.g., boric acid), burnable poison rods, and fuel assembly management.

PWR (VVER): A Pressurized Water Reactor of Russian design (***V***odo-***V***odyanoi ***E***nergetichesky ***R***eaktor, or "Water-water energetic reactor," i.e. water cooled and water moderated). VVER-440, -1000, and -1200 units produce 440, 1000, and 1200 MWe, respectively. The VVER-440 is an older technology, and the VVER-1200 is under development. Newer VVER units follow international safety standards and are sold to countries around the world.

SGHWR: Steam generating heavy water reactors are similar CANDU reactors, but use light water coolant to produce steam like a BWR.

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