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日本原子力研究開発機構

JAEA-Technology

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The amount of Pu in the spent fuel was evaluated from Xe isotopic ratio in off-gas in reprocessing facility, is related to burnup.

Six batches of dissolver off-gas (DOG) at spent fuel dissolution process were sampled from the main stack in Tokai Reprocessing Plant (TRP) during BWR fuel (approx. 30GWD/MTU) reprocessing campaign. Xenon isotopic ratio was determined with Gas Chromatography/Mass Spectrometry.

Burnup and generated amount of Pu were evaluated with Noble Gas Environmental Monitoring Application code (NOVA), developed by Los Alamos National Laboratory.

Inferred burnup evaluated by Xe isotopic measurements and NOVA were in good agreement with those of the declared burnup in the range from -3.8% to 7.1%. Also, the inferred amount of Pu in spent fuel was in good agreed with those of the declared amount of Pu calculated by ORIGEN code in the range from -0.9% to 4.7%.

The evaluation technique is applicable for both burnup credit to achieve efficient criticality safety control and a new measurement method for safeguards inspection.

Keywords: Xenon Isotopic Ratio, Off-gas, Burnup, Plutonium, Reprocessing Facility

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This work has been performed in JAEA as a joint research with Los Alamos National Laboratory.

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再処理オフガス中のキセノン同位体比測定による燃焼度及びプルトニウム量の評価技術
（共同研究）

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使用済燃料のせん断及び溶解時に発生するオフガス成分の1つであるキセノンの同位体比は、主として原子炉内での核反応の進行度に依存し、原子炉の型、燃焼度、プルトニウム生成量といった燃料の特性と相関を持つことが知られている。

このため、ロスアラモス研究所では、再処理施設から大気中に放出されたオフガス中のキセノン同位体比を測定することにより、燃料特性 (炉型、燃焼度、核種組成等)に関する情報を算出できる解析コード(NOble gas enVironmental monitoring Application)を開発してきた。キセノン同位体比測定とNOVAにより、処理した使用済燃料の炉型、燃焼度及びプルトニウム量を評価する技術が確立できれば、再処理施設の遠隔監視等が可能となり、保障措置技術の一つのオプションとして期待できる。しかしながら、再処理工程内のオフガス中のキセノン同位体比の実測データによるNOVAの検証はなされていない。

そこで本件では、東海再処理施設の溶解オフガス中のキセノン同位体比を測定し、NOVAを用いて、使用済燃料の燃焼度及びプルトニウム量の評価手法としての可能性を確認した。

測定したキセノン同位体比からNOVAを用いて使用済燃料の燃焼度及びプルトニウム量を評価した結果、BWR燃料であることが推定され、発電所側から示された燃焼度と-3.8%～7.1%で一致した。

一方、プルトニウム量については、燃焼度からORIGENコードを用いて計算した値と-0.9%～4.7%の差で一致した。このため、溶解オフガス中のキセノン同位体比測定により、使用済燃料の燃焼度及びプルトニウム量を把握することが可能であることが判った。

今後、各種オフガス中のキセノン同位体比を測定し、再処理施設における保障措置技術としての適用性を評価していく。

本研究は、日本原子力研究開発機構とロスアラモス研究所との共同研究に基づいて実施したものである。

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1. Introduction

The International Atomic Energy Agency (IAEA) is generally charged with the safeguarding of nuclear materials at reprocessing facilities. One particular challenge to IAEA safeguards is the monitoring of large scale reprocessing facilities devoted to civil uses.[1] Safeguarding these facilities and similar plants in the future will pose an interesting challenge to the IAEA safeguards system.

It is well-known that stable Xe isotopes produced with fission reflect spent fuel characteristics (fuel type, burnup, generated amount of Pu). Since dissolver off-gas, which is released with dissolution of spent fuel in reprocessing facility, contains most of the gaseous fission products, xenon isotopic ratios in the dissolver off-gas sampled from the stack would be expected to provide a new measurement method for application to safeguards inspection. Plutonium isotopic ratios and generated amount of Pu in the fuel were evaluated by using Xe isotopic ratio, which is related to burnup. The technique is also applicable for evaluating burnup credit to achieve efficient criticality safety control.

This report describes joint work between the Japan Atomic Energy Agency (former Japan Nuclear Cycle Development Institute) and Los Alamos National Laboratory (LANL) undertaken in Action Sheet-55: “Joint Studies of the Stable Fission Gas Verification Methodology.” This project has demonstrated the successful application of the stable noble gas isotope methodology to verification of nuclear material for safeguards.

2. Project Objectives

Action Sheet-55 describes joint work between JAEA and LANL. This project will demonstrate the application of the stable noble gas isotope methodology for verification of nuclear material for safeguards. The benefits of this methodology are (1) reduced intrusiveness; (2) lower costs compared to traditional methods of chemical analyses; (3) the ability to archive samples for later analyses, if necessary; and (4) an independent verifications compared to standard IAEA protocol. The technical goals of this demonstration project were to predict (1) fuel burnup, (2) ^{240}Pu/^{239}Pu ratio, (3) total Pu in the spent fuel, and (4) fuel type.

The project has three components: (1) on-stack sampling of the off-gas exhaust, (2) isotopic analysis of stable Xe isotopes in the off-gases, and (3) analysis of the isotopic data using the NOVA software to determine certain reactor parameters. JAEA had previously developed sampling protocols and has installed sampling ports on their off-gas exhaust systems. JAEA also had mass spectrometry equipment for the analysis of the off-gas xenon isotopes. Therefore, Action Sheet 55 was focused primarily on data analysis and interpretation.

3. Theory and Background Information

Proper development and implementation of a monitoring technique of the type proposed here requires knowledge of the nuclear fuel cycle. It is also necessary to understand the physics of fission product generation and depletion in a nuclear system. Brief descriptions follow.

The Nuclear Fuel Cycle and Routes to Nuclear Weapons

The nuclear fuel cycle (Fig. 1) comprises all operations from the mining of U ore to the final disposal of waste products. The “front-end” of the nuclear fuel cycle refers to the preparation of U for use in reactors. The “back-end” of the nuclear fuel cycle, which is of concern here, refers to operations performed on spent fuel (e.g., reprocessing and disposal).

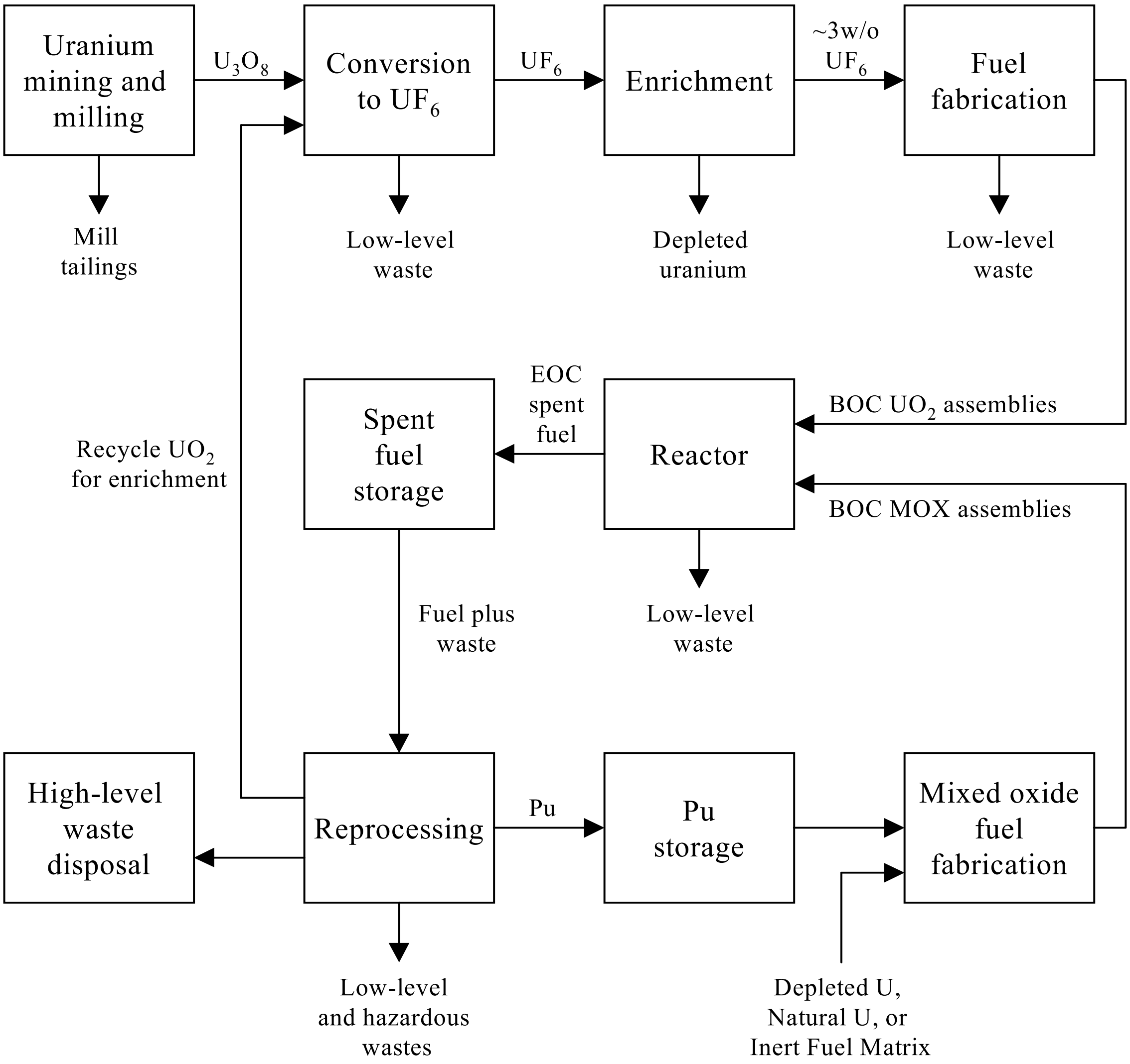

Figure 1 The nuclear fuel cycle with plutonium and uranium recycle.

The last step in the "front-end" portion of the nuclear fuel cycle is the fabrication of fuel elements and assemblies for use in a nuclear reactor. This process requires the conversion of the enriched UF_6 into solid pellets of UO_2. The pellets are inserted into metal tubes (called cladding) and sealed to form fuel elements. The elements are tested and bundled together into an assembly. The assemblies are then ready to be used in a nuclear reactor to produce heat and consequently electricity.

Depending on the reactor refueling schedule, the assemblies remain in the core for three to five years. During this time, some of U in the fuel is fissioned producing energy and fission products. Consequently, the discharged fuel is highly radioactive and requires significant shielding. The fuel is then transferred to a storage pool, where it is allowed to decay until the majority of the short-lived radioactive fission products decay to stable isotopes. While in the reactor, some of the neutrons are absorbed in the ^{238}U producing ^{239}Pu, after two subsequent β^- decays. Typical boiling water reactor (BWR) fuel with a burnup of 33,000 MWd/MTU contains about 96% U, 1% Pu, and 3% fission products. The majority of Pu in discharged fuel is composed of ^{239}Pu which is useful in weapons and reactor fuel. The ^{235}U concentration in the fuel is decreased from ~3% to slightly less than 1%; however, there is still a significant amount of ^{235}U remaining in the fuel.

Two options are available after the removal of the spent fuel from the spent fuel storage pools. The first option is permanent disposal, most likely in a geological depository. The second option is to attempt to recover the useable material from the spent fuel (i.e., Pu and U). Reprocessing refers to the procedure used to recover Pu and unburned U from the spent fuel. This material can then be refabricated into fuel rods and recycled into the reactor to provide more energy. Another advantage of reprocessing is that it leads to a decrease in the volume of the high-level waste. However, reprocessing also creates a potential proliferation hazard and therefore is of interest to the IAEA.

All reprocessing operations consist of four basic steps: (1) mechanically chop the spent fuel into small pieces, (2) dissolve the fuel in nitric acid, (3) use solvent extraction to separate the products of interest (U and Pu) and the waste into streams, and (4) dispose of the waste products. In the past, several reprocessing methods were developed including the REDOX, BUTEX, and PUREX processes. As far as it is publicly known, all reprocessing plants in the world employ some variation of the PUREX (Pu-U-Extraction) process.

The PUREX process consists of three cycles of solvent extraction using tri-n-butyl phosphate (TBP). PUREX uses liquid-liquid extraction principles and oxidation-reduction. The first step in the PUREX process is the mechanical disassembly of the fuel assembly into small pieces (called chopping). The fuel is then dissolved in nitric acid. During the chopping and dissolving phases, gases (such as ^3H, Kr, Xe, I_2, CO_2, NO_X, and steam) are released. These gases are transferred to a gas-treatment system for treatment, release, and/or recycle (nitrogen oxides can be converted back to nitric acid).

The fission product gases released during the chop and dissolve phases include noble gases such as Xe and Kr.

The chemically inert Xe and Kr generally travel directly to the stack and are relatively unaffected by chemical separations and porous filters. These gases (being fission products) contain information about the fuel being reprocessed and may prove a valuable monitor of reprocessing activities. Also, since the gases are emitted through the facility's stack, the best collection point for taking samples (i.e., on-stack) is relatively far away from the primary reprocessing activities. Other potential fission product monitoring points might be the high- and low-level waste streams and solvent streams; but because of their complex chemical and radioactive nature and their invasive sampling requirements, these are less attractive than the stack noble gases.

Noble Gas Production in Spent Nuclear Fuel

An ideal monitor for fuel burnup would have a large, consistent fission yield for all fissioning isotopes, would not vary with neutron energy, would not be destroyed or produced by absorption, would be stable, would not migrate in the fuel, and would be determinable with high accuracy and precision. Because such a monitor does not exist, it is crucial that any safeguards system consider the production, destruction, and decay of the monitor isotope in the spent fuel. In a typical nuclear fission reaction a variety of reaction products are generated including fission products, neutrons, gamma-rays, beta particles, and neutrinos. Also, a considerable amount of energy (usually on the order of 200 MeV per fission) is released. The fission products generated tend to be neutron-rich and are generally unstable. These products then decay over time to stable nuclei.

On average, two fission products of roughly equal mass are generated per fission; however, the frequency of the occurrence of symmetric fission products is rare. As can be seen from Figs. 2 and 3 (with fission yields from ENDF/B-VI), fission products generally cluster around two peaks with respect to mass number. The Xe and Kr isotopes tend to be located on or near these peaks; however, the fission yields for Kr isotopes are usually between 5 to 10 times lower than those for the Xe isotopes. Figures 2 and 3 also show the significant changes in fission product yields that occur with changes in the energy of the fission-inducing neutron and change in the fissioning isotope. It is these changes in fission yield which add to the system specific information (i.e., fuel type and burnup) contained in the fission product noble gases.

Properties of Various Xenon and Krypton Isotopes

Xenon and krypton have several properties that make them attractive for use as environmental monitors. Xenon (and to a slightly lesser degree Kr) are prevalent in spent fuel (see Figs. 2 and 3), yielding a large potential signal for measurement. Also, the relative concentrations of Xe and Kr isotopes change significantly with changes in fissioning isotope and neutron spectra; thus, the relative concentrations of these noble gas isotopes are indicative of various spent fuel parameters (including burnup, fuel type, operational history, etc.). The noble gas isotopes are chemically inert and therefore are relatively unaffected by the complex chemical processes that are involved in separating U and Pu from spent fuel. In addition, they are difficult and costly to remove from the stack gases. Thus, they are usually released directly through the stack.

Another characteristic of Xe and Kr isotopes that prove attractive for safeguards purposes is their low concentration in natural air. Generally speaking, background air contains approximately 87 ppb Xe and 1140 ppb Kr.[2] This implies that even low signatures of Xe and Kr released from a facility could be detected with a reasonably high degree of accuracy if sampled directly on-stack. Figures 4 and 5 contain plots of the percent isotopic compositions of Xe and Kr isotopes in natural air and in the gas produced directly from fission (the fission yields are taken from ENDF/B-VI). Note that the fission values are for ^{235}U thermal fission and do not include burnup and production due to neutron absorption in the fission products themselves or decay across mass chains (e.g., delayed neutron activity). One can note that the relative concentration of various Xe and Kr isotopes in natural air is markedly different from that produced in the gaseous elements from fission.

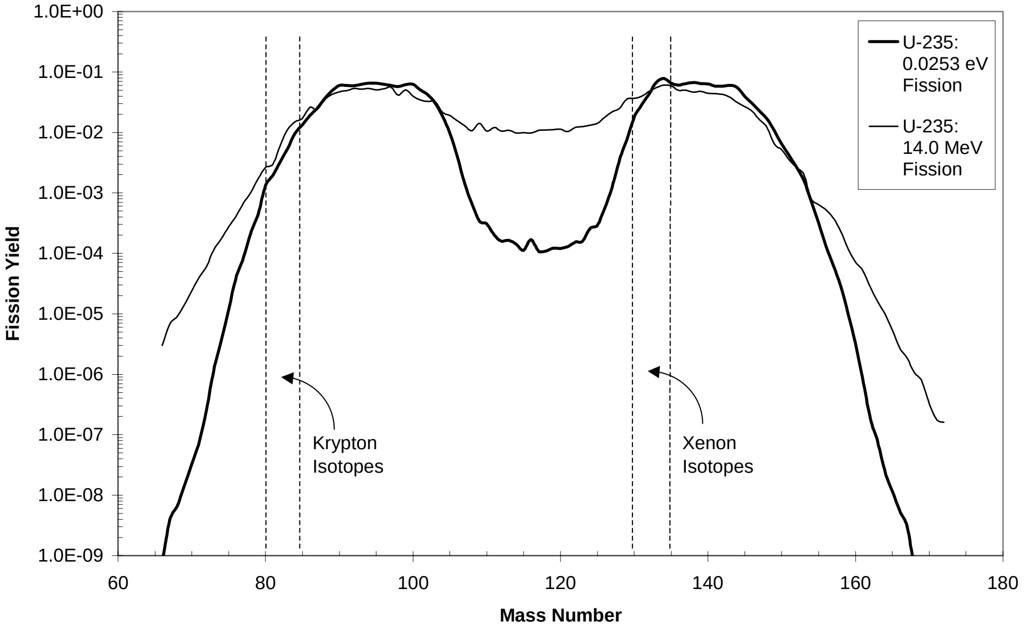

Figure 2 ^{235}U thermal and 14 MeV fission yields versus mass number.

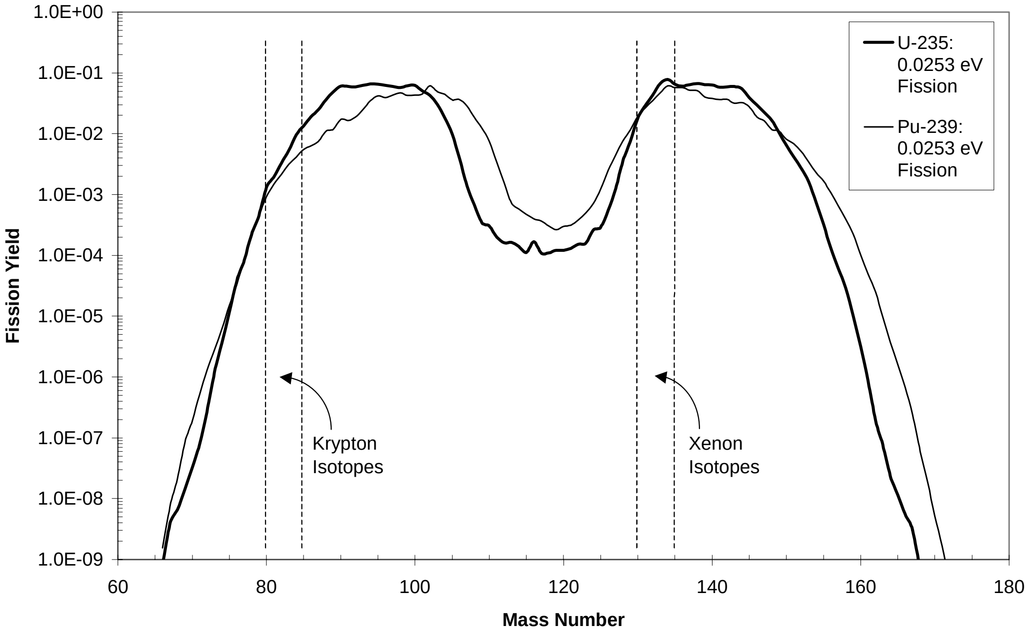

Figure 3 ^{235}U and ^{239}Pu thermal fission yields versus mass number.

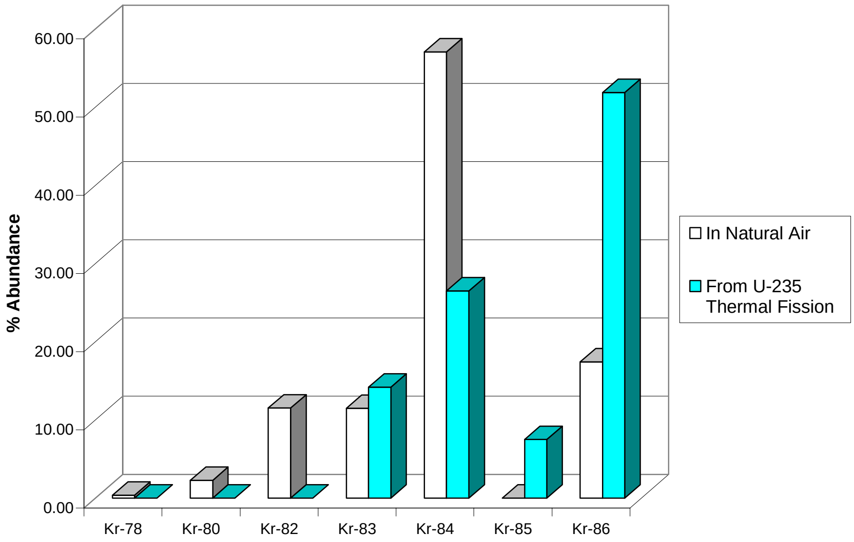

Figure 4 Percent isotopic abundance of various Kr isotopes in natural air and from ^{235}U thermal fission.

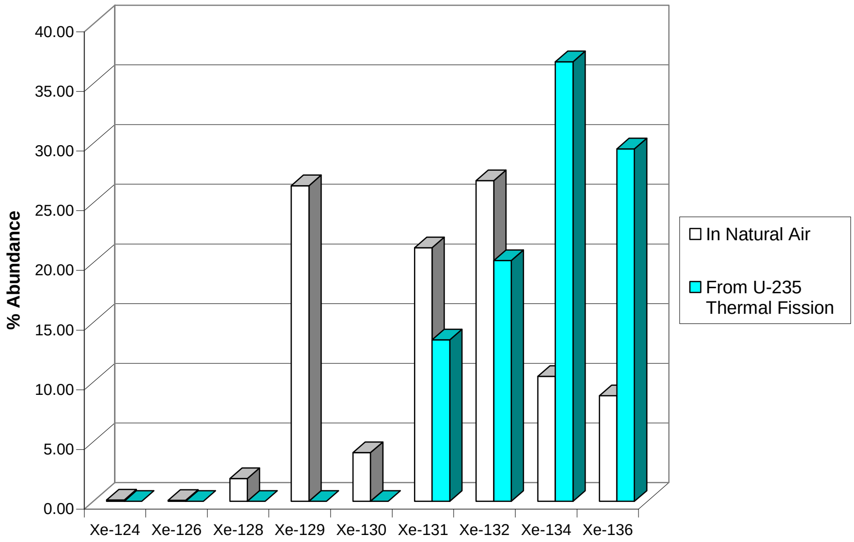

Figure 5 Percent isotopic abundance of various Xe isotopes in natural air and from ^{235}U thermal fission.

To aid in burnup determination, it is desirable to have a monitor that is invariant under all conditions (i.e., for each fission of any nuclide at any energy a specific probability of generation of this fission product exists). The monitor (fission product) should also not depend on reactor type, operational history, or power level (i.e., have short-lived parents and a small neutron absorption cross section). Since there is no single fission products with these characteristics, several fission products with some of these traits will be used together to derive the desired burnup information. Using several fission products together may also allow the determination of other spent fuel parameters (including fuel type, age, operational history, etc.).

In any on-stack measurement of noble gases, a mixture of the fissiogenic gas and natural air will be sampled. To aid in removing the background (or natural air) component, it will be necessary to have a measurement for a noble gas isotope that is not produced during fission. Below is an examination of Xe and Kr isotopes of interest, discussing the most appropriate isotopes for use in removing the background air component and the particular traits and properties of each isotope.

Figures 6 and 7 illustrate the production and destruction modes (fission, absorption, and decay) for Kr and Xe isotopes of interest. Note that all short half-life parents (i.e., less than 1 hour) have been assumed to decay instantaneously. The primary isotopes of interest from a safeguards standpoint are the stable isotopes (with the exception of ^{85}Kr), since all shorter-lived radioactive species will have significantly decayed prior to reprocessing. The fissioning process tends to create neutron-rich (unstable) fission products. The majority of stable Xe and Kr isotopes are then produced by the β^- decay of parent isotopes. The direct fission yields for stable isotopes are fairly small. Thus, in some cases there exists an appreciable time delay from fission to the generation of stable Xe and Kr isotopes (this may be ignored for the very short half-life parents). Note there is also some linkage between decay chains through neutron absorption. These factors (absorption, production from fission, and decay) and their differing effects for each isotope result in considerable information concerning the spent fuel being contained in the relative amounts of various Xe and Kr isotopes.

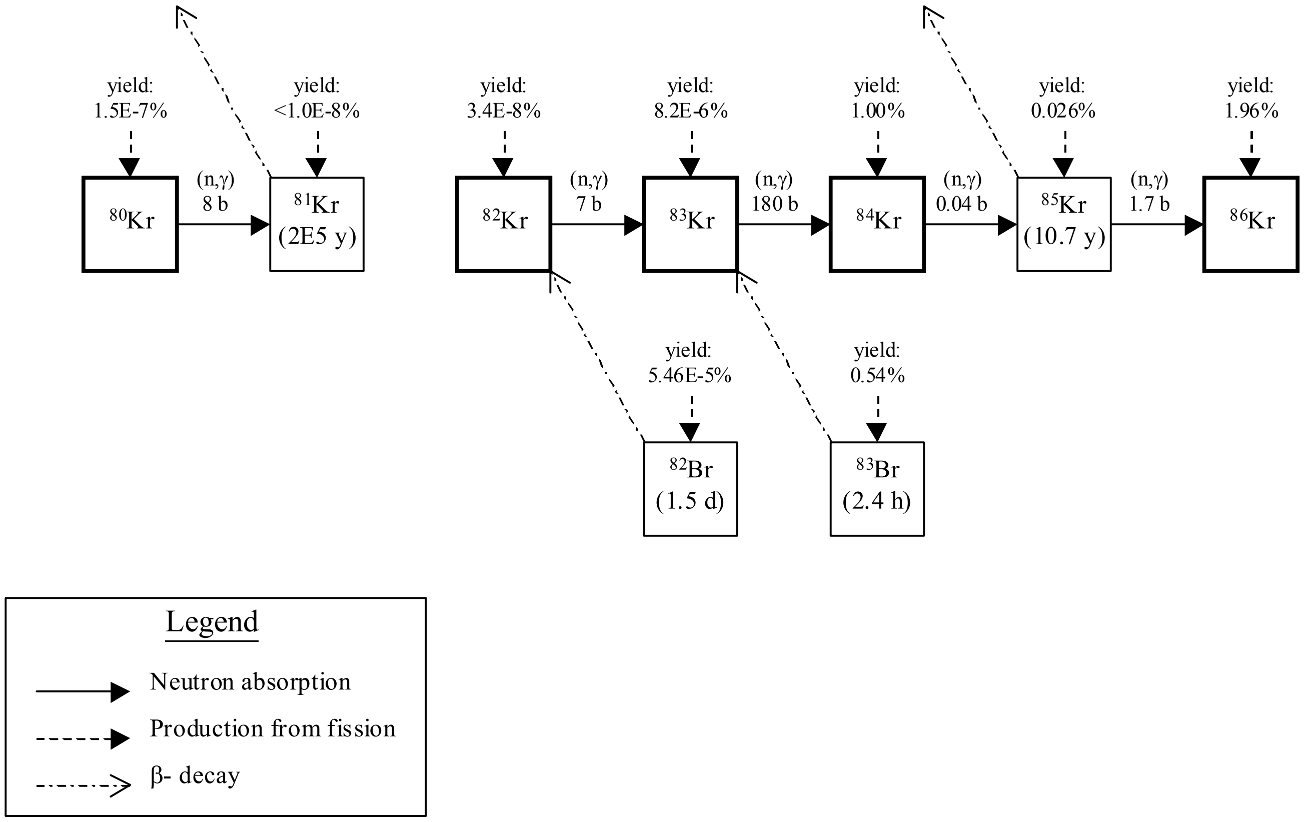

Figure 6 Krypton isotope production and decay chains (information from ENDF/B-VI and Ref. 3).

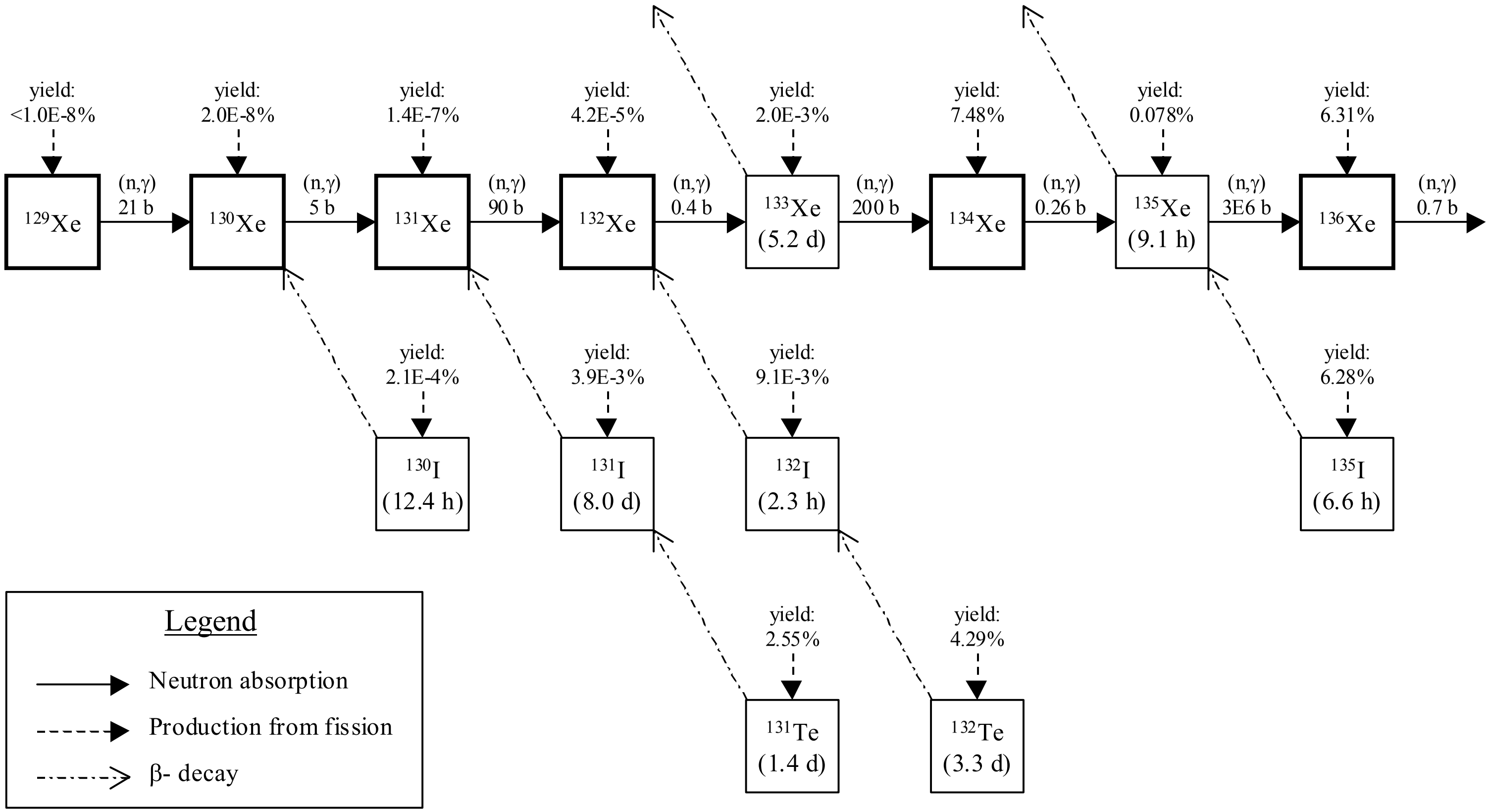

Figure 7 Xenon isotope production and decay chains (information from ENDF/B-VI and Ref. 3).

Krypton-80 and Krypton-81 are produced from parents with short half-lives. They can therefore be assumed to be produced directly from fission. ^{80}Kr is a stable isotope, and ^{81}Kr is radioactive with a 2.0×10^5 year half-life. Both of these isotopes have fission yields many orders of magnitude lower than the other interesting Kr isotopes. For this reason, it will be assumed that these isotopes are not produced in significant quantities via fission. Thus, ^{80}Kr is a candidate for removing the background air component. ^{81}Kr (which does not occur naturally) will not be used in this study.

^{82}Kr is produced from the decay of ^{82}Br. The cumulative yield for ^{82}Kr is small (several orders of magnitude lower than the higher mass Kr isotopes). The production and decay rates via neutron absorption for ^{82}Kr are also small. This implies that the ^{82}Kr concentration in the spent fuel is fairly invariant with respect to many reactor parameters including power level and operational history. This would make ^{82}Kr an ideal isotope for use as a burnup monitor; however, its small fission yield will make it difficult to measure. Thus, ^{82}Kr is a second candidate that may be used to aid in the removal of the background air component.

The larger fission yield (0.54%) and the shorter half-lives of its parent nuclides makes ^{83}Kr a good candidate as a burnup and fuel type monitor. The only drawback for this isotope is its large thermal neutron absorption cross section (180 b). Similar characteristics are found for ^{84}Kr. It has a good fission yield (1.00%) and extremely short-lived parent nuclides. Again its primary drawbacks are its production from neutron absorption in ^{83}Kr and its large abundance in natural air (see in Fig. 4).

^{85}Kr is a unique isotope in that it has short-lived parents, it has a small absorption cross section, and it is not naturally occurring; however, this nuclide is radioactive with a half-life of 10.73 years. ^{85}Kr also has a fairly small fission yield. Due to its radioactive nature, it is not useful to monitor burnup or to determine fuel type since its concentration in the spent fuel is dependent on the decay time since the fuel's removal from the reactor. For this reason, the relative isotopic concentrations of ^{85}Kr may be used to determine the spent fuel's age since discharge.

The last of Kr isotopes of interest is ^{86}Kr. This nuclide has a large fission yield (1.96%), short-lived parents, and a very small absorption cross section. It also has a small natural air concentration (see Fig. 4). This nuclide is extremely invariant with respect to the reactor's operational characteristics. Its concentration does not change with changes in power level, operational history, or decay time. Essentially, the relative concentration of ^{86}Kr in the spent fuel is an excellent measure of the burnup and fuel type.

An analysis of Figs. 5 and 7 shows that ^{129}Xe has an extremely small fission yield, is stable, and appears in large quantities in natural air. ^{129}Xe has a fairly large thermal neutron absorption cross section (21 barn); however, due to the size of its absorption cross section and the magnitude of the typical neutron flux in thermal reactors, neutron absorption does not significantly affect its concentration in spent fuel. Due to its small fission yield, it is appropriate to assume that ^{129}Xe is not produced in significant quantities by fission.

Thus, ^{129}Xe is a candidate isotope for use in removing the background air component from samples of stack gases.

^{130}Xe has only one relatively short-lived parent nuclide (^{130}I) and has small production and destruction modes from neutron absorption. Thus, the relative concentration of ^{130}Xe in the spent fuel is not going to be significantly affected by changes in power level or operational history. However, due to its low fission yield this isotope may not be produced in large enough quantities to be accurately measured.

The isotopes ^{131}Xe and ^{132}Xe are both produced in large quantities via fission (2.55% and 4.29%, respectively) and both have parent nuclides with long half-lives (on the order of days). Also, due to the large absorption cross section of ^{131}Xe, significant linkage exists between the two mass chains. Both of these isotopes will contain information regarding fuel type and burnup, but they will also have a slight dependence on power level and operational history. The actual extent of this dependence will be examined in more detail later.

The most invariant of Xe isotopes of interest is ^{134}Xe. This isotope has a large fission yield (7.48%), no long-lived parent isotopes, and a small absorption cross section. For these reasons, the concentration of ^{134}Xe in the spent fuel will depend only on the fuel type and burnup.

The last Xe isotope of interest is ^{136}Xe. This isotope has a large fission yield (6.31%), no long-lived parent nuclides, and an extremely small absorption cross section (0.7 barn); however, it is worth noting that the ^{136}Xe concentration is significantly affected by the ^{135}Xe neutron absorption. ^{135}Xe has an extraordinarily large thermal absorption cross section (2.6×10^6 barn), and both it and its parent nuclide (^{135}I) have both reasonably long half-lives (on the order of several hours) and large fission yields. For this reason, the power level and operational history of the reactor will have a strong affect on the ^{136}Xe concentration in the spent fuel. The effect is probably so large as to render this isotope useless for burnup and fuel determination; however, it may be possible to use the ^{136}Xe concentration in the fuel to determine information about the reactor power level and operational history. This will be examined in more detail later.

Low-Burnup Versus High-Burnup Fuel

One of the primary objectives of this project is to design a monitoring system to allow the operators or inspectors to determine if the fuel being reprocessed has a low-burnup or a high-burnup. The reason for the distinction between low-burnup and high-burnup fuels is primarily due to its relationship to Pu isotopics in the fuel. As the fuel is consumed in the reactor, absorptions in ^{238}U produced ^{239}Pu (after two subsequent β^- decays). If the fuel is allowed to reach higher burnups more of ^{239}Pu is converted into the higher mass Pu isotopes (especially non-fissile ^{240}Pu and ^{242}Pu). These higher mass Pu isotopes tend to be neutron poisons and make the potentially separated Pu less attractive as a weapons material.

Generally speaking, one of the most notable parameters in determining the quality of the separated Pu is the ^{240}Pu/^{239}Pu isotopic ratio. Figure 8 shows the variation of the ^{240}Pu/^{239}Pu isotopic ratio as a function of burnup for a U.S. PWR. Weapons-grade Pu is usually considered to have a ^{240}Pu/^{239}Pu isotopic ratio around 7%. In Fig. 8, the ^{240}Pu/^{239}Pu isotopic ratio increases sharply with burnup and approaches a value of between 50 to 80% at high burnups.

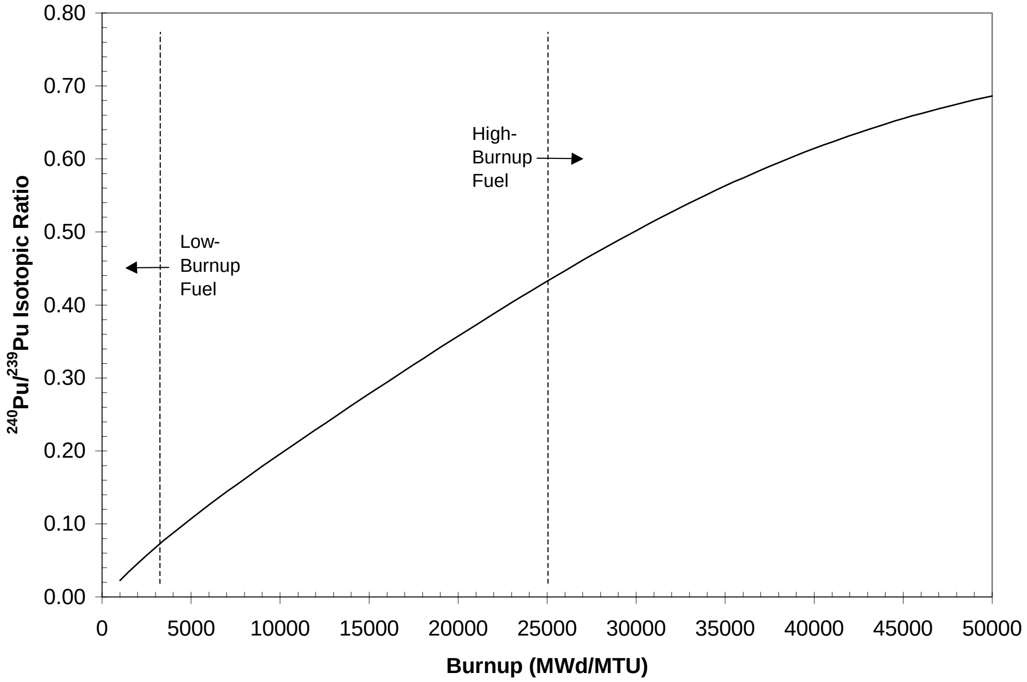

Figure 8 ^{240}Pu/^{239}Pu isotopic ratio versus burnup for a U.S. PWR calculated using the HELIOS[1] lattice physics code.

It would be desirable to have a monitoring technique that could be used to derive the following information about reprocessed spent fuel (in order of importance):

- Distinguish low burnup from high burnup fuels,
- Determine the spent fuel burnup,
- Determine the reactor type which produced the fuel,
- Determine other fuel parameters (including age, history, etc.),
- Determine ^{240}Pu/^{239}Pu isotopic ratio of the fuel,
- Determine Pu content of the fuel.

With these characteristics an inspector would have the capability to confirm the information declared by the reprocessing facility.

4. Previous Efforts

The concept of using fission products to determine spent nuclear fuel parameters (such as fuel type and burnup) dates back many years. In 1965, work was performed by W. J. Maeck to determine nuclear fuel burnup based on the ratio of two stable fission product isotopes of same element.[4] In this work, Maeck was interested in developing a method for accurately determining the burnup of spent fuel for fuel performance evaluations. He proposed using three isotopic ratios: ^{84}Kr/^{83}Kr, ^{132}Xe/^{131}Xe, and ^{144}Nd/^{143}Nd. These ratios could be used to obtain fairly accurate burnup, but required a significant amount of knowledge about the spent fuel including the reactor type and beginning-of-life (BOL) isotopics.

A comprehensive study of isotope correlation techniques (ICTs) was performed by L. Koch *et al.* at the European Transuranium Institute starting in 1970.[5-9] These studies focused on determining spent fuel parameters using heavy metal and fission product correlations derived from measured values. The studies relied heavily on destructive examination of the fuel and analysis of Cs and Nd isotopes in addition to the Xe and Kr isotopes. Xenon and Kr isotopic ratios analyzed were ^{84}Kr/^{83}Kr and ^{132}Xe/^{131}Xe. Although these studies suggested that there was promise in noble gas analysis, the experimenters were unsuccessful in generating a complete system for use at reprocessing sites based only on noble gas samples. Partly this was due to the fact that the correlations developed were based solely on measured values and were therefore only applicable to the individual reactor systems studied and were not extendable to cover a variety of different reactor types.

The European Safeguards Research and Development Association (ESARDA) directed by C. Foggi also investigated ICTs for analyzing spent fuel at reprocessing facilities.[10-12] Again their work was heavily focused on heavy metal and radioactive fission products, but some effort analyzed the use of stable Xe and Kr noble gases. One significant outcome of this study was its increased emphasis on reactor modeling, whereas in the earlier studies all of the correlations were based solely on measurements from one or two systems. The results of this study also showed that significant improvements in nuclear data were needed for the noble gas isotopes to be applied effectively on a wide scale.

A study published in 1988 by M. Ohkubo demonstrated the feasibility of using Xe and Kr gas for enhancing safeguards at reprocessing facilities.[13] The isotopic ratios ^{84}Kr/^{86}Kr, ^{86}Kr/^{83}Kr, ^{84}Kr/^{83}Kr, ^{132}Xe/^{131}Xe, ^{134}Xe/^{131}Xe, and ^{132}Xe/^{134}Xe were all used in the study. The calculations performed by M. Ohkubo for determining fission product concentrations versus burnup were simple; and therefore, the models used may have been in error. M. Ohkubo concluded that the technique was feasible; however, better models, more experimental data, and superior measurement techniques were necessary before it would prove effective at reprocessing facilities.

The use of noble gases as a monitor of proliferation activities was studied briefly at Idaho National Engineering Laboratory by T. C. Chapman.[14] The studies were preliminary in nature. One conclusion of these studies was that Xe fission isotopes were easier to measure and contained more information than Kr isotopes. They proposed a measurement technique which included separation of Xe gases from the air diluent and described a method for removing the background air contaminant; however, a completely integrated and validated system was not developed.

In 1993, G. B. Hudson at Lawrence Livermore National Laboratory performed a study analyzing the prospects of using Xe and Kr fission gases for monitoring reprocessing activities.[15] This study concentrated on determining the burnup and Pu composition of spent fuel using correlations derived from simple cross section models. Also, a significant literature search was performed to uncover numerous spent fuel measurements that included mass spectrometric measurements of Xe and Kr isotopes. Hudson concluded that it was feasible to use the noble gases, but that superior reactor models and better measurements were needed.

In the late 1990's, a study was performed by Y. Aregbe *et al.* that considered the monitoring of stable Xe and Kr gases at reprocessing facilities.[16-19] This study made significant improvements in measurement techniques applied and treated in detail the atmospheric dilution problem. Aregbe used KORIGEN code[20] to model PWR and CANDU reactors, which represented a notable enhancement to previous simple models and measurement correlations. In Aregbe's study, the techniques developed were not tested on "real" samples (i.e., taken from a reprocessing facility), rather synthetic samples of known isotopics were manufactured to test the measurement system. Though it was an improvement over previous attempts, Agrebe's study did not result in a practical system nor did the researchers consider broader application to other reactor types.

Most recently, researchers at Los Alamos National Laboratory (LANL) initiated a study to explore the feasibility of using stable Xe and Kr noble gases for use in monitoring reprocessing facilities.[21-22] These studies were mostly exploratory in nature and suffered from the use of overly simplified reactor models. However, one major improvement in this work performed was the use of advanced data analysis techniques in determining burnup and reactor types from measured values.

Though significant effort has been spent exploring the feasibility of environmental monitoring by stable noble gas measurements, before this project an integrated system using sophisticated data analysis techniques, state-of-the-art measurement systems, and rigorous reactor analysis methodologies which could be used at a reprocessing facility did not exist. The objective of this work was to produce such a system and to validate it for use with several fuel types.

5. Monitoring System Overview

The verification technique developed in this project combines on-line sampling, an isotope ratio mass spectrometer, and an accurate fissiogenic gas database through the use of a sophisticated data analysis technique. The complete integrated system is illustrated in Fig. 9. Development of this verification technique centered around three primary sections: the measurement system, the reactor physics calculations, and the data analysis methodology.

The measurements begin with the collection of gas samples at Tokai Reprocessing Plant (TRP) during the chopping and/or dissolution of spent fuel. The gas samples are then analyzed with a mass spectrometer capable of determining Xe and Kr isotopic ratios in the sample. The measurement system for on-stack analysis requires fast sample processing and the ability to aid in the removal of background air.

The reactor physics database contains Xe and Kr fissiogenic isotopic ratios and Pu concentrations as a function of burnup for an exhaustive set of fuel types. These ratios and Pu concentrations were calculated using a series of state-of-the-art reactor analysis codes. The codes used in developing this database allowed for accurate calculations of Pu, Xe, and Kr concentrations in spent fuel for a variety of reactor types. To properly couple the database to the measured isotopic ratios, the reactor analysis codes were benchmarked for the production of Xe, Kr, and Pu in as many reactor types as data allowed.

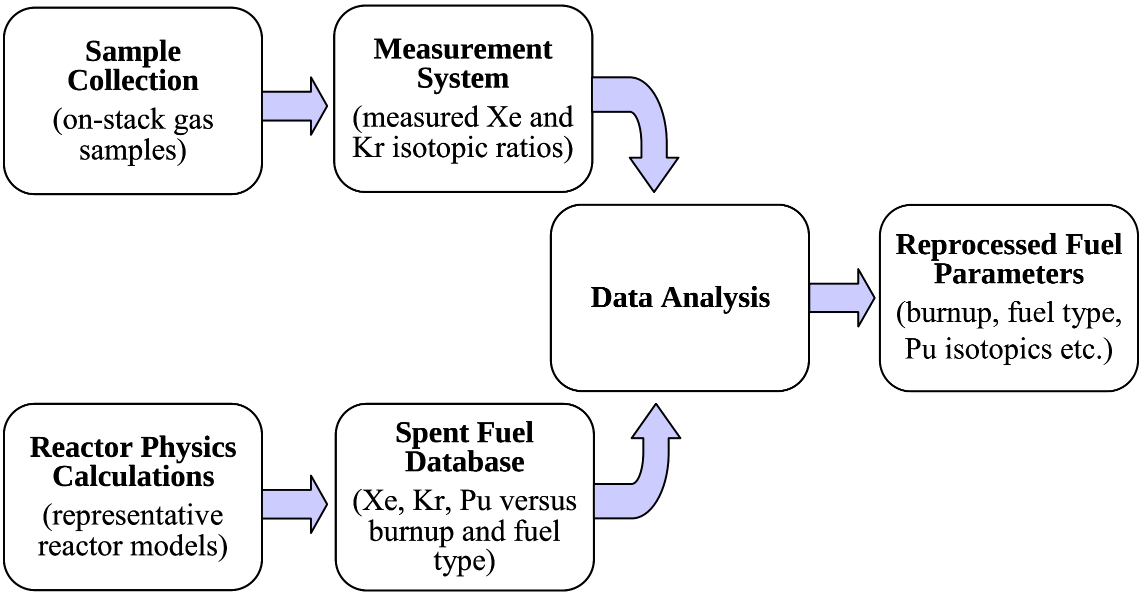

Figure 9 Verification technique overview.

The measured isotopic ratios were coupled to the reactor physics database using a sophisticated data analysis technique allowing the determination of fuel type [e.g., PWR, BWR, CANDU, etc.], fuel burnup, and Pu composition. This data analysis procedure included a method for removing any background air contamination resulting from dilution of the fission gas in natural air. The data analysis technique determined the most likely fuel type and burnup to match the measured isotopic ratios. Also, a resulting measure of the confidence in the result based on the uncertainties in the measured isotopic ratios and the calculated database is generated.

The analysis made use of each isotope's increased dependency on various fuel parameters to increase the versatility and capability of the verification technique. The most valuable isotopic ratios for use in determining burnup and fuel type were ^{131}Xe/^{134}Xe, ^{132}Xe/^{134}Xe, ^{83}Kr/^{86}Kr, and ^{84}Kr/^{86}Kr. ^{134}Xe and ^{86}Kr were chosen as the normalizing isotopes due to their larger fission yields and limited dependence on operational parameters. Additional isotopic ratios, that have much smaller fissiogenic components (e.g., ^{130}Xe/^{134}Xe and ^{82}Kr/^{86}Kr), may still prove useful in these analyses. Also, since the ^{135}Xe neutron absorption cross section is so large (~2.6×10^6 barns), the ^{136}Xe/^{134}Xe isotopic ratio contains information regarding the operational history of the fuel and may be used to determine factors such as power level and percent downtime. The radioactive nature of ^{85}Kr made the ^{85}Kr/^{86}Kr ratio ideal for determining the spent fuel age (i.e., the time from discharge).

The technique developed here was applied to solve two separate but similar problems: the Inverse Problem and the Forward Problem. The Inverse Problem occurs when an inspector takes an air sample from a reprocessing facility and, without any other previous information, analyzes it to determine the fuel type, burnup, and other properties of the spent fuel. The Forward Problem is slightly simpler in that the inspector assumes that he knows the fuel type being reprocessed (perhaps declared by an operator or observed by the inspector) and simply uses the air sample to determine fuel burnup and isotopic composition.

Both problems use the same basic technique (i.e., measurement system, calculated reactor physics information, and Bayesian data analysis); however, they require two separate reactor physics databases. The reactor physics database for the Inverse Problem consisted of an exhaustive set of reactor models. Each model of which was a good representative average of all the reactors of that type (i.e., the representative PWR model was a good average of a large set of different PWRs). The database for the Forward Problem contained calculated values for a large set of specific fuels (e.g., Westinghouse 17×17 PWR with 3.00 w/o ^{235}U fuel or GE 8x8 BWR with 2.50 w/o ^{235}U fuel). This usage of two different databases allowed for the determination of information at two different levels. For the Inverse Problem, the information determined (i.e., burnup, fuel type, and Pu composition) was generally less accurate than that from the Forward Problem; however, the Inverse Problem did not require information from the operator.

The usage of these two problems gives the inspector added capabilities. If the inspector's aim is simply to verify declared information from an operator, then the Inverse Problem will allow that inspector to determine the burnup and fuel type independent of any information supplied by the operator. If the inspector desires to have an accurate accounting of Pu concentrations in the fuel, then the Forward Problem allows the inspector the ability to accurately predict Pu isotopic composition of the fuel; however, this may rely upon the usage of information supplied by the reprocessing facility. The usage of the solutions to these two problems together and separately is discussed in more detail later.

The final product of this project was a user-friendly application that yields burnup, fuel type, fuel age, and Pu isotopics using measured Xe and/or Kr isotopic ratios. The tool is capable of analyzing both the Forward and Inverse Problems. This tool is accurate and versatile in determining fuel parameters. Also, the tool is easy enough to use that little training is required for its proper application.

6. Reactor Physics Database

One of the key components to the NOVA code is an extensive reactor physics database that determines the relationship between measured Xe and Kr isotopic ratios and spent fuel parameters (such as burnup and Pu concentrations).[23] The reactor physics database contains Xe and Kr fissiogenic isotopic ratios and Pu concentrations as a function of burnup for an exhaustive set of fuel types. These ratios and Pu concentrations were calculated using a series of state-of-the-art reactor analysis codes including HELIOS and Monteburns.[24, 25] To properly couple the database to the measured isotopic ratios, the reactor-analysis codes were benchmarked for the production of Xe and Pu in 12 different reactor types.

7. Data Analysis (NOVA)

The measured xenon isotopes are analyzed using the NOVA code. NOVA is a Visual Basic code that incorporates calculated reactor physics databases and a Bayesian analysis procedure to allow for the determination of various spent fuel parameters (including fuel type, burnup, fuel age, and Pu concentrations) from isotopic ratio measurements. This code allows the user to input any number of measured isotopic ratios (background corrected or not background corrected), performs the correction for any background-air contamination (if necessary), analyzes the ratios for either an Inverse or Forward Problem (as specified by the user), and outputs the solution (including expected Pu and fission product concentrations in the spent fuel). NOVA has numerous options available including the ability to selectively remove certain fuel types from the analysis and the ability to use any of a number of different reactor physics databases.

The measured isotopic ratios are coupled to the calculated fissiogenic gas database through a Bayesian analysis technique that allows for the determination of the most likely Pu concentrations and burnup from a set of measured isotopic ratios. Because any realistically acquired samples contain both a fissiogenic component and a natural-air component, the sample's measured isotopic ratios will consist of a combination of the fissiogenic and atmospheric-air noble gases. Because some noble gas isotopes (e.g., ^{129}Xe) are not produced in significant quantities via fission, these measured non-fissiogenic isotopes can be used to remove the background-air contaminant. This requires using known natural abundances of Xe isotopes in air (either assumed or measured). For Xe, with normalizing isotope ^{134}Xe and non-fissiogenic isotope ^{129}Xe, the isotopic ratio of interest is given by

$$\frac{N_{f,u}^{x}}{N_{f,u}^{134}} = \frac{\left(\dfrac{N_{m,u}^{x}}{N_{m,u}^{129}} - \dfrac{N_{m,air}^{x}}{N_{m,air}^{129}}\right)}{\left(\dfrac{N_{m,u}^{134}}{N_{m,u}^{129}} - \dfrac{N_{m,air}^{134}}{N_{m,air}^{129}}\right)}. \qquad (1)$$

Thus, given a measurement of the isotope of interest and the normalizing isotope (^{134}Xe) relative to ^{129}Xe in the unknown sample and in atmospheric-air, the background-air contaminant can be removed directly.

Given a set of I measured isotopic ratios [$R^m = (R_1{}^m, R_2{}^m, ...R_I{}^m)$], their associated standard deviations ($\sigma_i{}^m$), and a mutually exclusive, exhaustive set of J reactor models [$M = (M_1, M_2, ...M_J)$], we can determine the most likely model (M_j) at a particular burnup (B_j) using a Bayesian analysis methodology. The reactor models are described by a database of calculated isotopic ratios [$R_{ij}{}^c = (R_{ij1}{}^c, R_{ij2}{}^c, ...R_{ijK}{}^c)$] and their associated standard deviations ($\sigma_{ijk}{}^c$) for each model (M_j) at a series of K burnup points [$B_j{}^c = (B_{j1}{}^c, B_{j2}{}^c, ...B_{jK}{}^c)$]. The model based probabilities for each isotopic ratio [i.e., the probability that one would measure the isotopic ratio ($R_i{}^m$) given spent fuel from the reactor model (M_j) at burnup (B_j) and any background information (E)] can be calculated using the maximum entropy formulation:

$$p(R_i^m \mid M_j, B_j, E) = \frac{1}{\sigma_{ij}\sqrt{2\pi}} \exp\left[-\frac{(R_i^m - R_{ij}^{\mu})^2}{2(\sigma_{ij})^2}\right]. \qquad (2)$$

Using these model-based probabilities and a version of Bayes' theorem, the probability that the spent fuel is from reactor model (M_j) at burnup (B_j) given a set of measured isotopic ratios (R^m) can be determined from

$$p(M_j, B_j \mid R^m, E) = \frac{p(M_j, B_j \mid E)\prod_{i=1}^{I} p(R_i^m \mid M_j, B_j, E)}{\sum_{j=1}^{J}\prod_{i=1}^{I} p(R_i^m \mid M_j, B_j, E)\, p(M_j, B_j \mid E)}. \qquad (3)$$

The quantity $p(M_j,B_j|E)$, called the prior, represents the probability that the fuel is of type M_j at a burnup of B_j given any background evidence alone.

8. Experimental

Dissolver Off-Gas

The outline of the dissolver off-gas (DOG) treatment process and sampling point is shown in Figure 10. Off-gases from three dissolvers pass through a condenser, acid absorber, caustic washing column and HEPA filter at TRP. All sampled DOG was generated from BWR fuel assemblies which were reprocessed at TRP. Another off-gas sample was collected as background during rinsing of the dissolver with nitric acid.

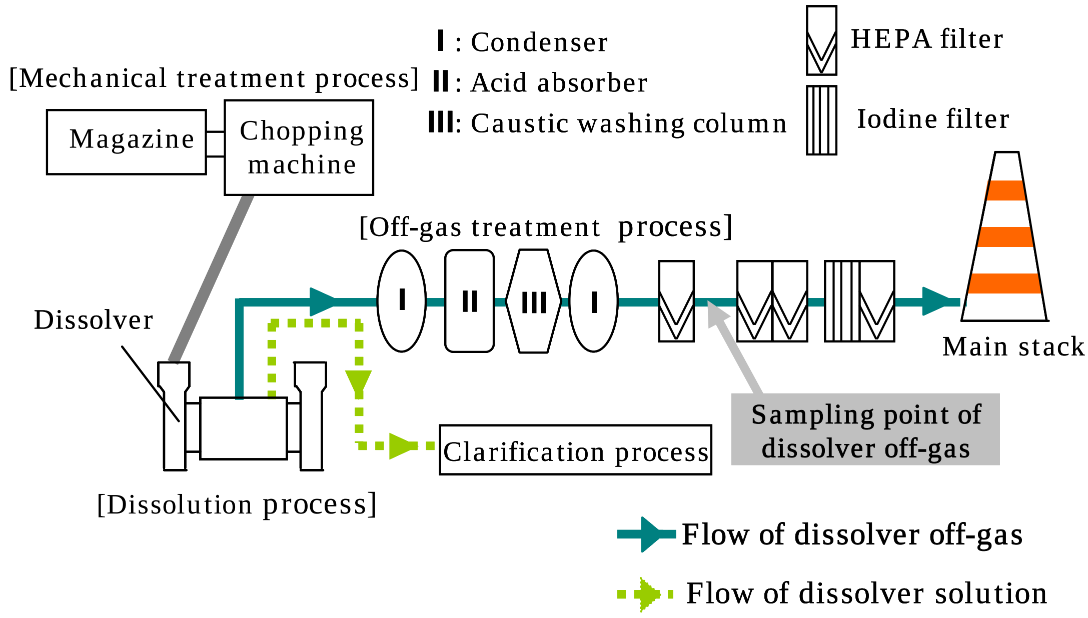

Figure 10 Outline of dissolver off-gas process.

The specification of the spent fuel for each of the 6 DOG samples is shown in Table 1; the sampling schedule is shown in Table 2. Because two assemblies of BWR fuel were dissolved after chopping into pieces about 5cm long, the burnup is the average of that of two assemblies.

Table 1 Specification of spent-fuel response to sampled dissolver off-gas

DOG* sample	Burnup (MWD/MTU)		Specific power (MW/MTU)	Cooling days	Initial U amount (Kg)	Enrichment of ^{235}U (%)
BWR-A	29,305	29,305	18.80	3,877	172.658	2.9
		29,305	18.80	3,877	172.406	2.9
BWR-B	28,586	28,106	18.80	3,878	172.651	2.9
		29,065	18.80	3,878	172.415	2.9
BWR-C	28,970	25,008	18.80	4,663	184.201	2.6
		32,931	20.10	2,805	172.684	3.1
BWR-D	27,992	25,174	18.80	6,469	184.620	2.6
		30,810	20.10	6,469	172.950	3.1
BWR-E	28,437	23,188	18.80	2,811	184.379	2.6
		33,686	20.10	2,811	172.066	3.1
BWR-F	28,298	25,093	18.80	4,670	183.931	2.6
		31,502	20.10	2,812	172.855	3.1

*DOG; Dissolver Off-Gas

Table 2 Sampling schedule of dissolver off-gas

DOG* sample	Dissolution time	Sampling time (Passing time**)
BWR-A	19:00～5:05	20:15～20:20 (1.25h)
BWR-B	6:15～16:00	10:03～10:09 (3.80h)
BWR-C	14:45～0:00	17:25～17:32 (2.67h)
BWR-D	15:30～0:45	17:45～17:52 (2.25h)
BWR-E	7:45～17:45	10:27～10:35 (2.70h)
BWR-F	7:15～17:00	10:19～10:25 (3.07h)
Background	―	13:56～14:01

*DOG; Dissolver Off-Gas

**After beginning dissolution

Table 3 shows timing of processes during normal operation of the dissolver at TRP. Spent-fuel, except covering material such as Zircaloy, is dissolved in 3M nitric acid at 100 degrees centigrade for 8 hours. During dissolution, oxygen is introduced to the dissolver to oxidize NOx which originate in the dissolution. Sampling of DOG was carried out from 1 hour to 4 hours after beginning of dissolution. Figure 11 shows the gamma-ray intensities near sampling point of DOG which is related to the concentration of radioactive elements in DOG. Gamma-ray intensity reached its peak after 90 minutes, coming back down the background level after 2 hours.

Table 3 Time table for normal operation of the dissolver for dissolution of spent fuel

0 – 30 min.	30-60 min.	60 – 90 min.	90-150 min.	150-480 min.
Supply HNO_3	Heat（~50℃)	Hold（~50℃)	Heat (~100℃)	Hold (100℃) Add HNO_3

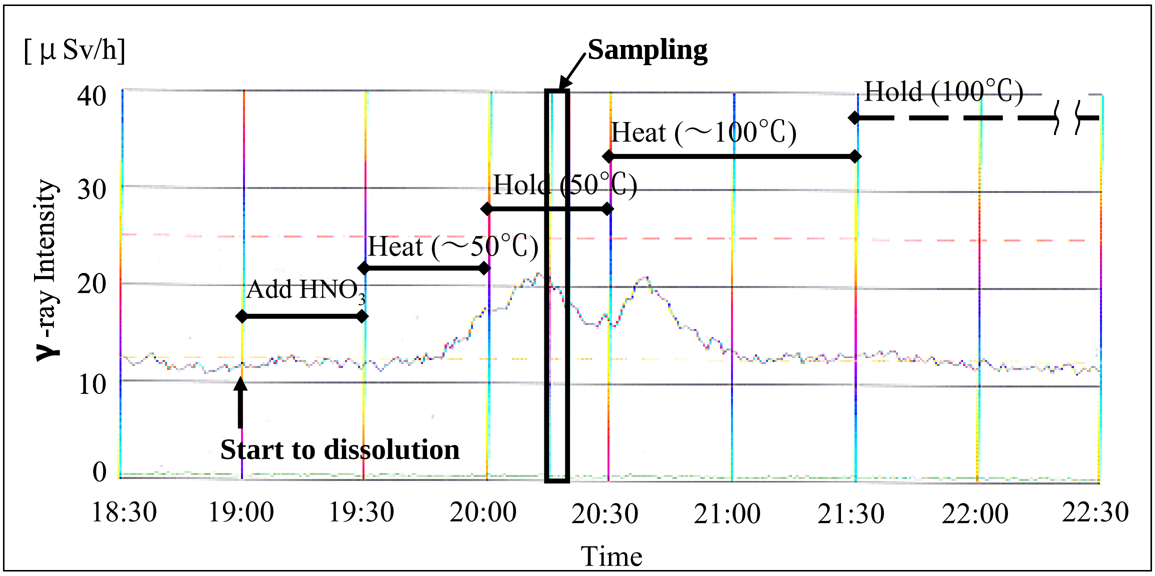

Figure 11 Gamma ray intensity near flow duct of dissolver off-gas during dissolution of BWR-A.

Sampling System of DOG

Figure 12 shows the DOG sampling system in hood box at TRP. The sampling system consists of compressor, vacuum pump, and sampling bottle, and it is equipped with a by-pass line for gas purging, valves to prevent back-flow, and gauges for pressure measurement. The sampling system is described by the specifications in Table 4 and by the schematic diagram of sampling system shown in Figure 13.
The DOG sampling system is evacuated before gas collection. During sampling, about 7 liter of DOG is compressed into a 1 liter sampling bottle. It takes 30 minutes to collect DOG including operation for vacuum, DOG purge, and purification of the line by air purge.

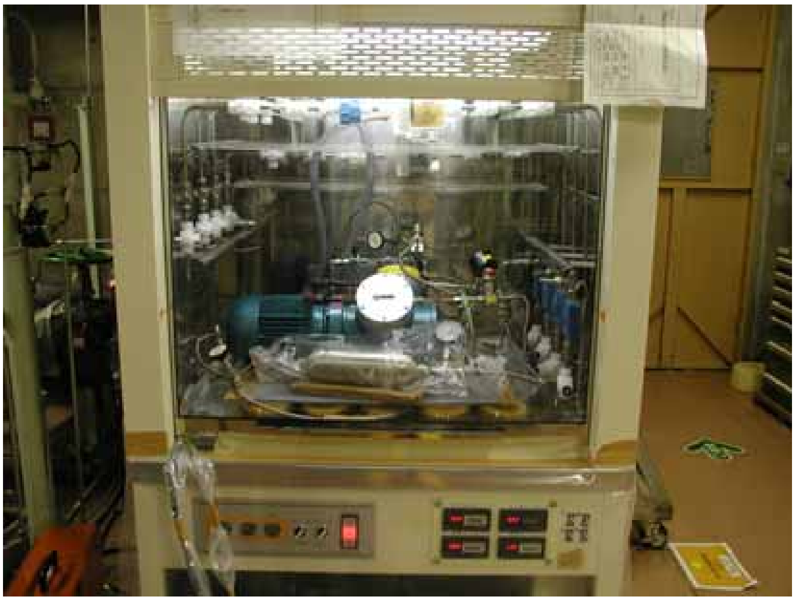

Figure 12 Sampling system of dissolver off-gas.

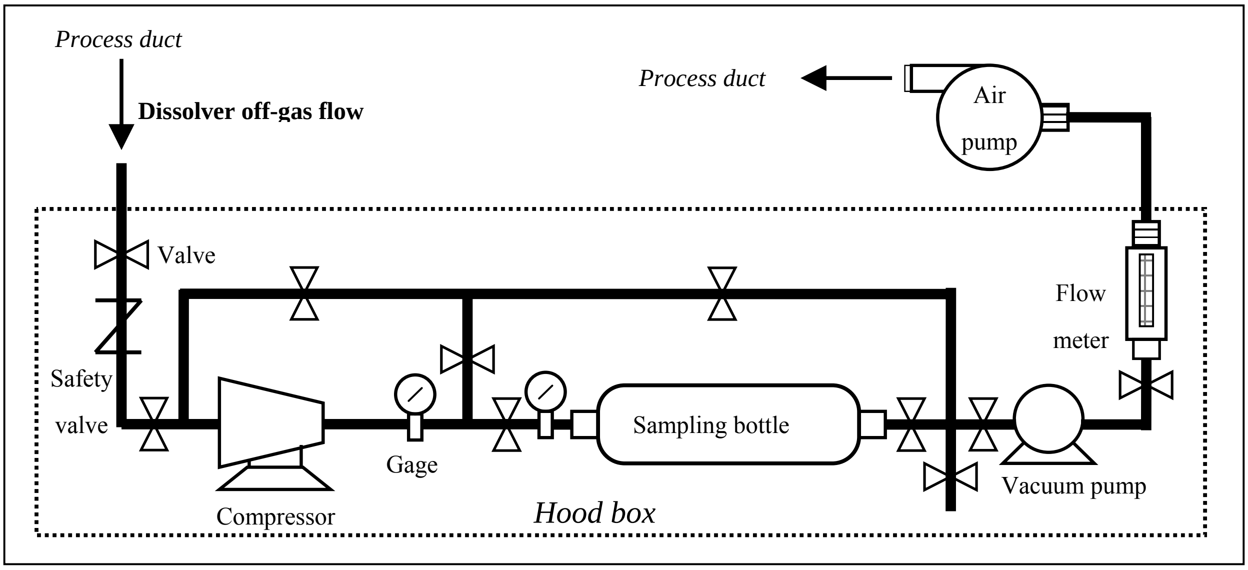

Figure 13 Schematic diagram of sampling system of dissolver off-gas.

Table 4 Specification of sampling system of dissolver off-gas

Item(model)	Company	Specification
Compressor (PJ15822-286.13)	KNF Co.	2L/min (In: atmosphere, Out: 0.7MPa)
Vacuum pump (DAM-010)	ULVAC Co.	7L/min, max. 0.4kPa
Sampling bottle	Swagelok Co.	D.O.T bottle, 1 liter, SUS304
Filter (SS-4FW-15)	Swagelok Co.	SUS316, 70µm, 15µm
Tube、Connecter	Swagelok Co.	SUS316
Valve	Swagelok Co.	
Gage	MKS Co.	-0.1~1.6MPa

Gas Cromatograph/Mass Spectrometer

Specifications and picture of gas chromatograph/quadrupole mass spectrometer (GC/MS) used to measure Xe isotope abundances in DOG is shown in Table 5 and Figure 14, respectively. This GC/MS was demonstrated to be suitable for the determination for low concentrations (ppm) of Xe and Kr.

Table 5 Specification of GC/MS

Model (Manufacturer): MASS I (Nichiden Aneruba Company)	
GC Unit	Column: MS5A (130 °C) Carrier gas: He (Purity: >99.999%, Pressure: 200 kPa, Velocity: 10mL/min)
MS Unit	Analysis tube: NAG-539 mass filter (Quadrupole type) Turbo molecule pump: TPH/U510 Hakutou company Rotary pump: 2030H Ionization energy: 17eV
Common	Dehumidifier: DHE-112 Komatsu electronics company Control system: YHP-9815S Gas pressure controller: Baratron type 222A. MKS company. Page gas: N_2 (Purity : >99.999%, 490 kPa) Thermal converter: 1.5 °C Detection Limit: Kr:0.1ppm (^{84}Kr, S/N＝3:1), Xe:0.1ppm (^{129}Xe, S/N=3:1), CH_4:0.05ppm (S/N＝3:1) Precision and Accuracy: ±5% ± 0.05ppm (≤1000ppm Xe) Stability: ±5%±0.05ppm/h, ±10% ± 0.05ppm/8h

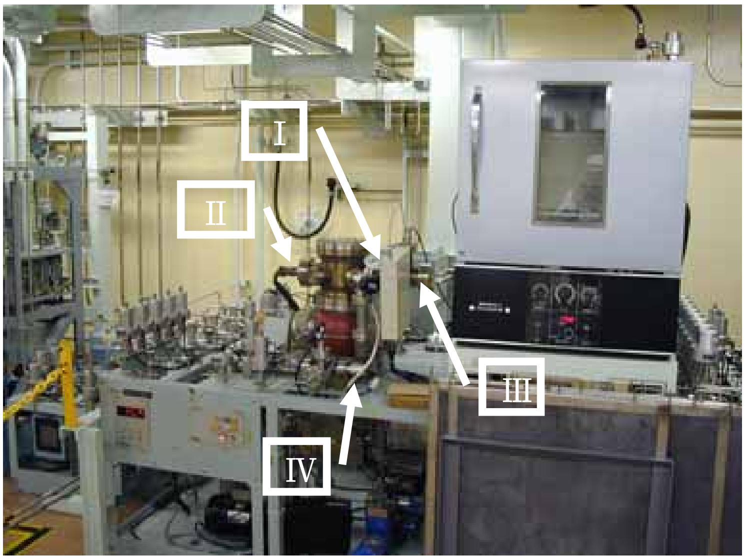

Figure 14 Gas chromatograph/mass spectrometer: MASS I.

I: Secondary electron multiplier, II: Ion source, III: Quadrupole, IV: Turbo molecular pump

Xenon gas standard

In order to evaluate the precision and accuracy of GC/MS, a Xe in N_2 standard was prepared. The composition of Xe standard is shown in Table 6.

Table 6 Composition of xenon standard gas

Element	Concentration (ppm)	Ratio (wt%)
Xenon	99.1	—
^{124}Xe	0.099	0.10
^{126}Xe	0.089	0.09
^{128}Xe	1.9	1.92
^{129}Xe	26	26.3
^{130}Xe	4.1	4.14
^{131}Xe	21	21.2
^{132}Xe	27	27.3
^{134}Xe	10	10.1
^{136}Xe	8.8	8.89
Krypton	89.1	—
Methane	20.0	—
Nitrogen	Balance	—

9. Results and Discussion

Statistical analysis of GC/MS

Figure 15 shows a typical gas chromatogram of Xe isotopes in Xe standard gas measured with the GC/MS. Xenon isotopes were observed 9 minutes after introducing DOG to the column. Peak shapes after 11 minutes were unstable because of detector noise. However, integrated ion currents from 9 minutes to 11 minutes are proportional to concentration of Xe isotopes and were used in subsequent calculations.

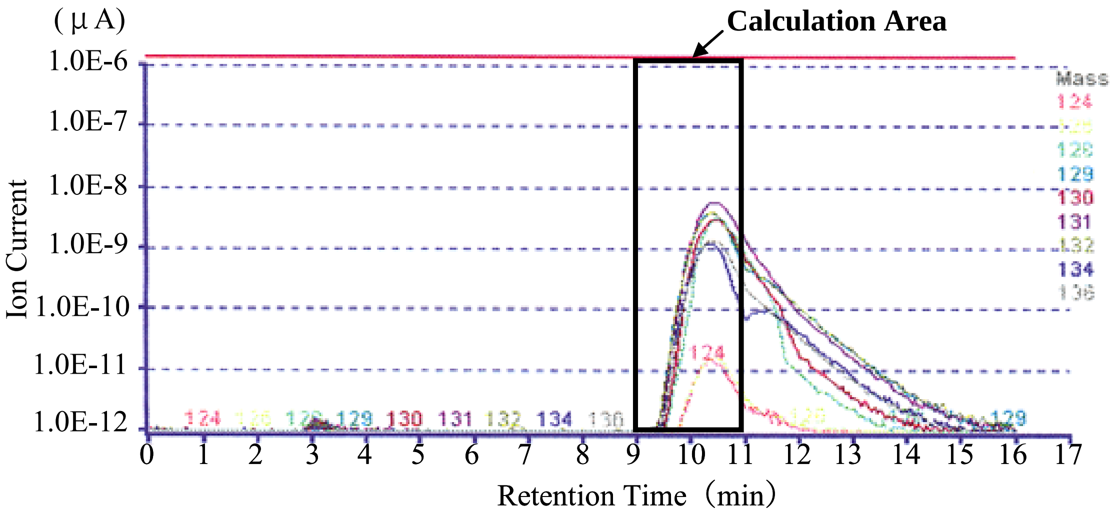

Figure 15 Gas chromatogram of xenon isotope in xenon standard gas.

The precision and accuracy of the GC/MS used in this study were characterized by multiple analysis of a standard containing 89.1 ppm Kr and 99.1ppm Xe in N_2 at 0.05 MPa (0.5 atm). The data are shown as relative abundance is shown in Table 7. These same data were used to calibrate the mass spectrometer. Xenon isotope concentration (ppm) was calculated as: Intensity $_{\text{(Standard)}}$ * [Ion Current $_{\text{(Sample-Background)}}$ / Pressure $_{\text{(Sample)}}$] / [Ion Current $_{\text{(Standard)}}$ / Pressure $_{\text{(Standard)}}$]. Acceptably reproduced data (<2.4 %RSD) were observed for all isotopes, and the %RSD for the key ratios ^{131}Xe/^{134}Xe and ^{132}/Xe/^{134}Xe were <2% RSD. Agreements between the measured and certified value were good except very low abundance (under 0.2 weight percent) isotopes such as ^{124}Xe and ^{126}Xe.

Table 7 Reproducibility of xenon isotope abundances in xenon standard gas determined by GC/MS

No.	Isotope Abundance (%)								
	^{124}Xe	^{126}Xe	^{128}Xe	^{129}Xe	^{130}Xe	^{131}Xe	^{132}Xe	^{134}Xe	^{136}Xe
1	0.119	0.114	1.97	27.7	4.15	20.9	26.4	10.3	8.36
2	0.121	0.116	1.96	27.6	4.06	21.2	26.5	10.2	8.38
3	0.126	0.112	1.98	26.9	4.22	21.2	26.8	10.2	8.45
4	0.125	0.115	1.99	26.7	4.23	21.5	26.9	10.0	8.43
5	0.121	0.113	1.98	27.3	4.12	21.2	26.5	10.3	8.36
6	0.126	0.114	1.94	27.1	4.09	21.3	26.6	10.3	8.42
7	0.123	0.118	1.98	26.6	4.26	21.4	27.0	10.0	8.49
8	0.125	0.122	2.01	27	4.21	21.2	26.6	10.2	8.47
9	0.122	0.117	1.97	27.2	4.19	21.2	26.5	10.3	8.40
10	0.126	0.116	1.96	27.2	4.21	21.2	26.5	10.3	8.39
11	0.124	0.114	1.91	27.3	4.19	21.2	26.6	10.3	8.18
12	0.122	0.117	1.94	27.2	4.25	21.4	26.7	10.0	8.33
Average (%)	0.123	0.116	1.97	27.1	4.18	21.2	26.6	10.2	8.39
Standard Deviation	0.0025	0.0027	0.027	0.318	0.066	0.150	0.172	0.112	0.082
Relative Standard Deviation (%)	2.01	2.34	1.38	1.17	1.58	0.71	0.7	1.10	0.98
Standard gas(%)	0.10	0.09	1.92	26.3	4.14	21.2	27.3	10.1	8.89
Average/ Standard gas (%)	20	33	2.6	3.2	1.0	0.2	-2.5	-1.0	-5.6
Nature (%)	0.10	0.09	1.91	26.40	4.10	21.20	26.90	10.40	8.90

Off-Gas Sample Analysis

The results of the analyses of the off-gas sampled from the stack during reprocessing are shown in Table 8. The dates of analysis are shown under each batch number. Data for minor isotopes ^{124}Xe and ^{126}Xe are not shown for clarity. Each sample and blank was analyzed twice.

Mass Spectrometer

The precision, expressed as the per cent relative standard deviation (%RSD) and accuracy of measurement are typical of that for a quadrupole mass spectrometer. The data also show a significant decrease in precision and accuracy for the minor isotopes ^{124}Xe and ^{126}Xe. This decrease is expected and not of consequence in these experiments since the more abundant isotopes ^{129}Xe, ^{130}Xe, ^{131}Xe^{132}Xe and ^{134}Xe are used to determine fuel type and burnup. The precision and accuracy demonstrated in the measurements of these isotopes is quite adequate in this application. Isotopic ratios were calculated from the signal intensities; the background correction is insignificant in that it would have little effect on the determination of spent fuel type and burnup.

Table 8 Results of xenon isotope abundances in dissolver off-gas determined by GC/MS

DOG* sample		Isotope Abundance (%)								
		^{124}Xe	^{126}Xe	^{128}Xe	^{129}Xe	^{130}Xe	^{131}Xe	^{132}Xe	^{134}Xe	^{136}Xe
BWR-A	n1	1.72E-03	5.44E-04	3.30E-02	3.20E-03	0.165	8.59	21.4	28.3	41.6
	n2	1.74E-03	5.46E-04	3.27E-02	3.16E-03	0.165	8.61	21.3	28.1	41.8
	Ave.	1.73E-03	5.45E-04	3.29E-02	3.18E-03	0.165	8.60	21.3	28.2	41.7
BWR-B	n1	5.78E-03	1.48E-03	3.39E-02	1.12E-02	0.159	9.25	20.9	27.8	41.8
	n2	5.77E-03	1.46E-03	3.25E-02	1.06E-02	0.153	8.89	21.0	27.7	42.2
	Ave.	5.78E-03	1.47E-03	3.32E-02	1.09E-02	0.156	9.07	20.9	27.8	42.0
BWR-C	n1	5.86E-03	1.51E-03	3.63E-02	1.19E-02	0.175	8.72	21.5	28.3	41.3
	n2	5.57E-03	1.44E-03	3.53E-02	1.20E-02	0.171	8.93	22.0	28.3	40.5
	Ave.	5.72E-03	1.48E-03	3.58E-02	1.19E-02	0.173	8.83	21.8	28.3	40.9
BWR-D	n1	1.16E-02	2.77E-03	3.68E-02	3.01E-02	0.154	8.72	21.1	28.9	41.0
	n2	0	2.48E-03	3.68E-02	2.68E-02	0.173	9.15	21.1	28.7	40.8
	Ave.	5.78E-03	2.63E-03	3.68E-02	2.84E-02	0.163	8.94	21.1	28.8	40.9
BWR-E	n1	2.11E-03	9.24E-04	3.55E-02	1.05E-02	0.160	8.98	21.2	28.1	41.4
	n2	2.20E-03	9.77E-04	3.59E-02	1.08E-02	0.162	8.69	21.4	28.1	41.6
	n3	2.26E-03	9.87E-04	3.61E-02	1.07E-02	0.161	9.10	21.3	27.8	41.6
	Ave.	2.19E-03	9.63E-04	3.58E-02	1.07E-02	0.161	8.93	21.3	28.0	41.5
BWR-F	n1	5.29E-03	1.91E-03	3.60E-02	1.51E-02	0.163	8.65	21.3	28.9	41.0
	n2	5.41E-03	2.13E-03	3.66E-02	1.51E-02	0.166	8.68	21.4	28.6	41.1
	Ave.	5.35E-03	2.02E-03	3.63E-02	1.51E-02	0.165	8.66	21.3	28.7	41.1
Blank	n1	0.48	0.22	0	13.4	2.63	15.4	24.2	19.2	24.5
	n2	0.52	1.83	0.16	16.4	2.66	17.4	24.7	16.6	19.6
	Ave.	0.50	1.02	0.08	14.9	2.64	16.4	24.5	17.9	22.0

*DOG; Dissolver Off-Gas

Determination of Spent Fuel Type and Burnup

Xenon isotopic ratios provided were analyzed using the LANL analysis capability to determine the spent fuel type and burnup. The following isotopic ratios were generated from the raw data: ^{130}Xe/^{129}Xe, ^{131}Xe/^{129}Xe, ^{132}Xe/^{129}Xe, ^{134}Xe/^{129}Xe, and ^{136}Xe/^{129}Xe. The same ratios were generated for the data labeled as “blank”. The “blank” data was used to subtract “background” air. The samples all contained very little background air contaminant (>99% fissiogenic xenon in the samples); thus, the background air subtraction was a minor step.

^{130}Xe/^{129}Xe, ^{131}Xe/^{129}Xe, ^{132}Xe/^{129}Xe, and ^{134}Xe/^{129}Xe isotopic ratios were used to infer a fuel type and burnup for each sample. Off-gas sample data from six batches (BWR-A, BWR-B, BWR-C, BWR-D, BWR-E, and BWR-F) were provided. Following background air subtraction, the following ratios were available for each sample: ^{130}Xe/^{134}Xe, ^{131}Xe/^{134}Xe, and ^{132}Xe/^{134}Xe. These ratios were used to infer the fuel type and burnup of the fuel containing these ratios of fissiogenic Xe. Using all three of these ratios, the fuel type was inferred to be BWR fuel for all samples. Using only the ^{131}Xe/^{134}Xe and ^{132}Xe/^{134}Xe ratios (the two strongest signals), a mixture of inferred fuel types was generated. Figure 16 shows the results of fuel type inference for all samples. The “Other” fuel type category includes MAGNOX fuel, LMFBR driver Fuel, LMFBR target Fuel, and High-Temperature Gas Reactor (HTGR) Fuel. As can be seen, the success rate for proper fuel type inference decreases significantly when less than three ratios are used.

Fuel burnup was also inferred from the ratios available (^{130}Xe/^{134}Xe, ^{131}Xe/^{134}Xe, and ^{132}Xe/^{134}Xe). Figure 17 shows the inferred fuel burnup versus the declared fuel burnup when using all three of these ratios. Figure 18 shows the inferred fuel burnup versus the declared fuel burnup using only the two strongest ratios (^{131}Xe/^{134}Xe and ^{132}Xe/^{134}Xe only). Again, significantly better agreement is found when using all three ratios than when using only two ratios.

Figure 19 shows the confidence level for the inferred fuel burnup and fuel type for each measured sample when using all three isotopic ratios. Generally good confidence is available in the results; however, there are a number of datapoints that appear to have significant uncertainties. These datapoints typically only result in a slight miscalculation in the fuel burnup or split decisions between PWR and BWR fuel types. It should be noted that only a slight difference in isotopic ratios is available between PWR and BWR fuel types and determining the fuel type difference between these two is often difficult (especially with only two ratios).

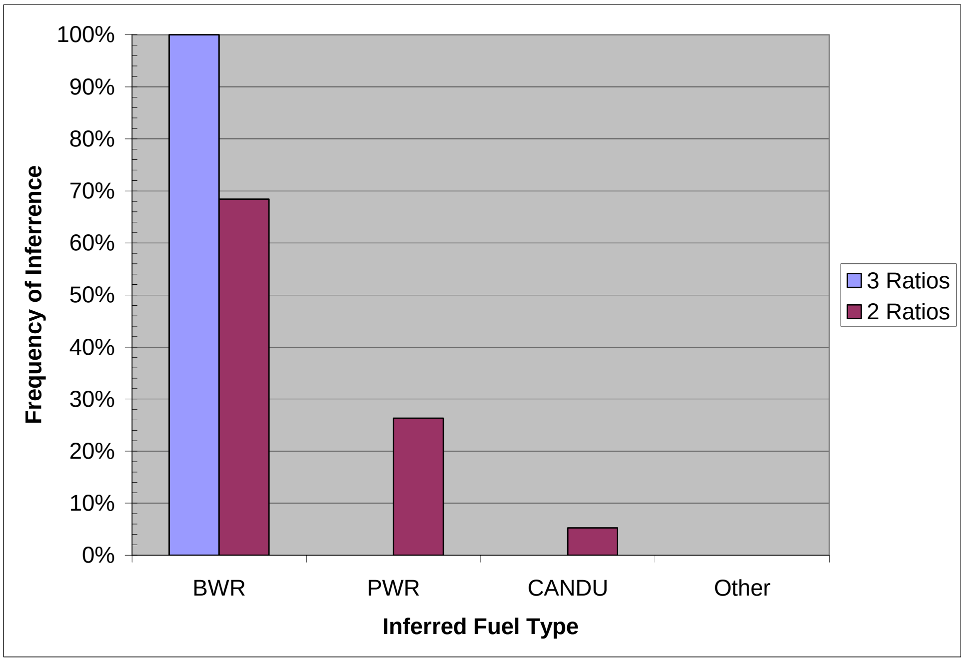

Figure 16 Frequency of fuel type inference when using three ratios (^{130}Xe/^{134}Xe, ^{131}Xe/^{134}Xe, and ^{132}Xe/^{134}Xe) and when using two ratios (^{131}Xe/^{134}Xe and ^{132}Xe/^{134}Xe).

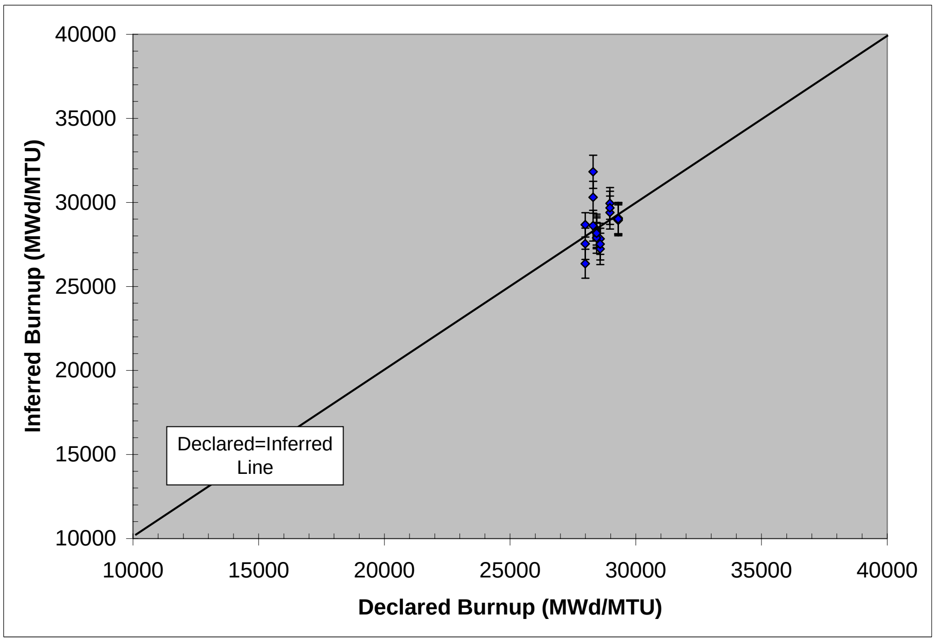

Figure 17 Inferred fuel burnup (based on ^{130}Xe/^{134}Xe, ^{131}Xe/^{134}Xe, and ^{132}Xe/^{134}Xe isotopic ratios) versus declared fuel burnup.

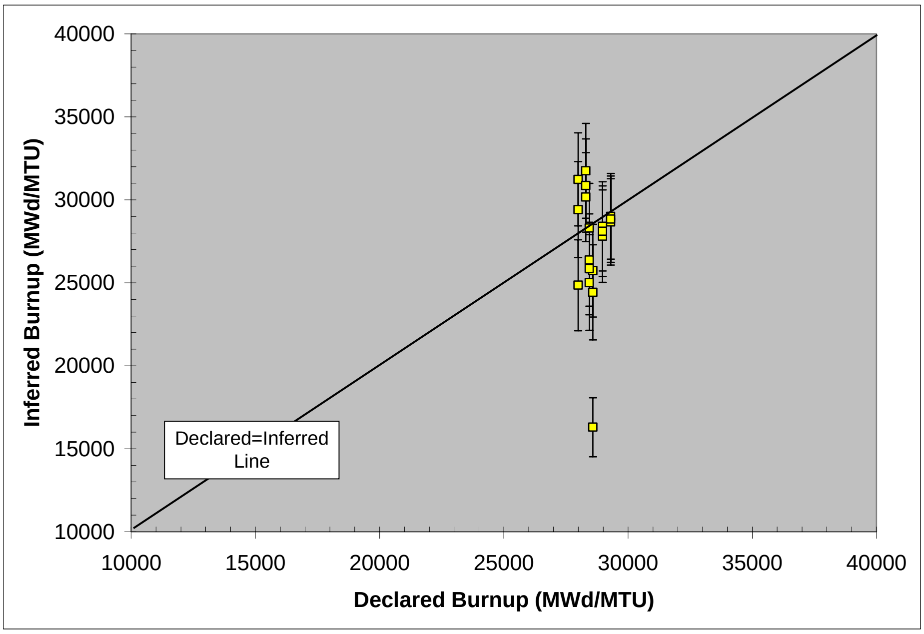

Figure 18 Inferred fuel burnup (based on $^{131}Xe/^{134}Xe$ and $^{132}Xe/^{134}Xe$ isotopic ratios) versus declared fuel burnup.

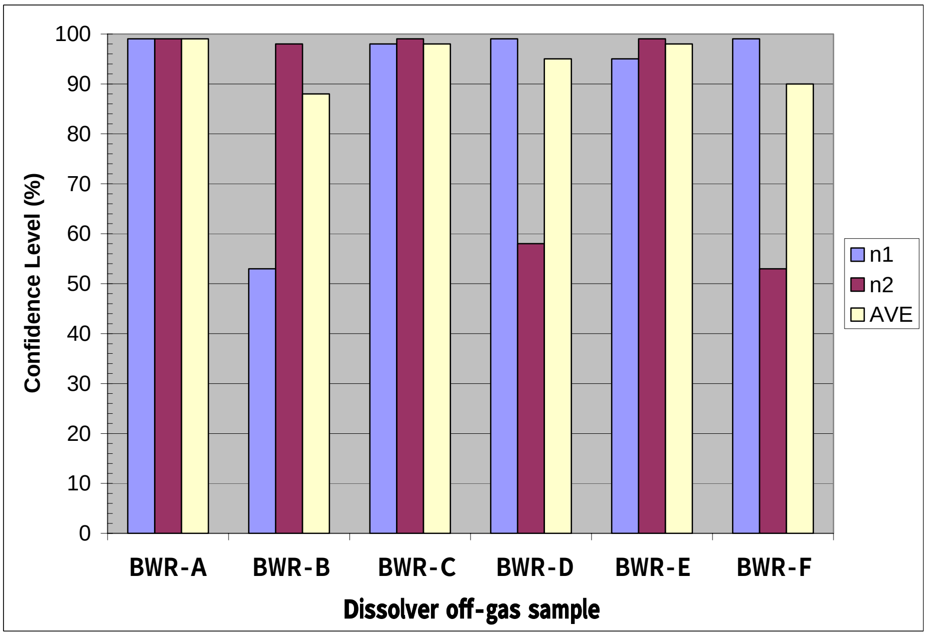

Figure 19 Result confidence level for the samples within each batch (based on result using $^{130}Xe/^{134}Xe$, $^{131}Xe/^{134}Xe$, and $^{132}Xe/^{134}Xe$ isotopic ratios).

Evaluation of Generated Amount of Plutonium

Plutonium concentration in spent fuel was calculated using the HELIOS-1.4 NOVA database. Using Xe isotopic ratios only Pu concentrations can be determined since the total quantity of material dissolved is not available in that data. Table 10 shows the NOVA Pu concentrations compared to the declared Pu concentrations calculated from the data in Table 9.

Table 9 Specification of spent-fuel sampled using dissolver off-gas.

DOG* sample	Burnup (MWD/MTU)		Cooled day	Charged		Post irradiated		
				U (Kg)	^{235}U (wt%)	U (Kg)	^{235}U (wt%)	Pu (g)
BWR-A	29,305	29,305	3,877	345.1	2.9	331.5	0.83	2,830
		29,305	3,877					
BWR-B	28,586	28,106	3,878	345.1	2.9	331.8	0.83	2,786
		29,065	3,878					
BWR-C	28,970	25,008	4,663	356.9	2.6	343.1	0.81	2,902
		32,931	2,805		3.1			
BWR-D	27,992	25,174	6,469	357.6	2.6	344.1	0.80	2,753
		30,810	6,469		3.1			
BWR-E	28,437	23,188	2,811	356.4	2.6	342.9	0.81	2,906
		33,686	2,811		3.1			
BWR-F	28,298	25,093	4,670	356.8	2.6	343.2	0.81	2,865
		31,502	2,812		3.1			

*DOG; Dissolver Off-Gas

Table 10 Plutonium results from Xe isotopic composition and concentration in dissolver off-gas

DOG* sample	Pu Concentration (kg/MTU)		
	Declared	NOVA	Percent Difference (%)
BWR-A	8.20	8.22	0.2
BWR-B	8.07	8.00	-0.9
BWR-C	8.13	8.31	2.2
BWR-D	7.70	8.01	4.0
BWR-E	8.15	8.10	0.7
BWR-F	8.03	8.41	4.7

*DOG; Dissolver Off-Gas

10. Conclusions and Recommendations

An accurate and non-invasive monitoring technique to support safeguards measurements at reprocessing facilities was demonstrated successfully by scientists at JAEA (former JNC), LANL, and the Texas A&M University. This technique involves the on-stack collection and measurement of isotopic ratios of stable noble fission gases from emissions during reprocessing of spent fuel using gas chromatograph/mass spectrometer. All the technologies used in these measurements are commercially available and relatively inexpensive. The isotopic ratios of selected Xe nuclides were compared to a database of calculated isotopic ratios using a data analysis method to determine specific fuel parameters (e.g., burnup, fuel type, fuel age, etc.). These inferred parameters could be used to verify operator declarations. The complete system has been integrated into a user-friendly software application (named NOVA). NOVA is a visual basic user interface coupling a Bayesian data analysis procedure to a calculated reactor physics database (produced using the Monteburns 3.01 code system and the HELIOS code system). The calculated database was well benchmarked for many reactor types. The complete system (GC/MS, reactor modeling, and data analysis) was validated using on-stack measurements during the reprocessing of low-burnup target fuel from a BWR production reactor at TRP. These measurements led to an inferred burnup that matched the declared burnup with sufficient accuracy and consistency for most safeguards applications.

In past work at LANL, the NOVA code was tested using numerous light water reactor measurements from the literature. NOVA was capable of accurately determining spent fuel type, burnup, and fuel age for these experimental results. NOVA was also capable of distinguishing between PWR and BWR reactors (esp. when using both Xe and Kr isotopic ratios). In all, NOVA is capable of determining all of the following characteristics for any reprocessed fuel:

- Distinguish low burnup from high-burnup levels,
- Determine the spent fuel burnup (within ~ 4%),
- Determine the reactor type with produced the fuel,
- Determine the fuel age (with ~ 4%),
- Determine ^{240}Pu/^{239}Pu isotopic ratio for the spent fuel,
- Determine Pu content of the spent fuel,
- Determine the concentration of various other fission products of interest in the spent fuel.

Each of these capabilities has been validated using measured data except for the determination of the concentrations of other fission products. But although the technique demonstrated in this project shows great promise, it does require further validation.

Further validation is necessary to demonstrate the robustness of this system for different fuel types. In this system only BWR fuels were analyzed. The existing reactor physics database has not been tested for other fuel types and any results for these types of reactors should be suspect prior to proper validation.

Simple experiments that would be valuable in future joint projects between JAEA and LANL would consist of first analyzing the measured data in an inverse problem to confirm the declared fuel type and burnup and the reanalyzing the measured ratios in a forward problem to aid in material control and accountancy of Pu in spent fuel. It would also be very useful to include Kr fission products in future trials of the NOVA software.

References

1. T. SHEA, S. DERON, F. FRANSSEN, D. HOPE, N. ISLAM, S. JOHNSON, E. KUHN, G. LASZLO, D. NEAL, and T. RENIS, “Safeguarding Reprocessing Plants: Principles, Past Experiences, Current Practices, and Future Trends,” *J.N.M.M.,* 21(4), p. 17 (1993).

2. M. OZIMA and F. A. PODOSEK, *Noble Gas Geochemistry*, p. 12-13, Cambridge University Press, Cambridge (1983).

3. F.W. WALKER, J.R. PARRINGTON, and F. FEINER, Nuclides and Isotopes: Fourteenth Edition, General Electric Company, San Jose (1989).

4. W. J. MAECK, “Proposed Determination of Nuclear Fuel Burnup Based on the Ratio of Two Stable Fission Products of the Same Element,” IDO-14642, Idaho Operations Office, U.S. Department of Energy (1965).

5. L. KOCH, H. BRAUN, and A. CRICCHIO, “Some Correlations Between Isotopes of Xe, Kr, U, Pu, and Burnup Parameters for Various Thermal and Fast Reactors,” *Proceedings of the Symposium on Progress in Safeguards Techniques*, Karlsruhe, West Germany, July 6-10, p. 539, International Atomic Energy Agency (1970).

6. L. KOCH, A. CRICCHIO, and F. GERIN, “Isotope Correlations of Heavy Isotopes and Fission Products for Consistency Checks and Data Generation,” presented at the IAEA Working Group on the Use of Isotopic Composition Data in Safeguards, Vienna, April 10-14, 1972.

7. R. WELLUM, R. DE MEESTER, K. KAMMERICHS, and L. KOCH, "The Categorization of Binary Isotope Correlations Derived from the Databank of the European Institute for Transuranium Elements," *Proceedings of the ESARDA Symposium on the Isotopic Correlation and Its Application to the Nuclear Fuel Cycle*, Stresa, Italy, May 2-6, European Safeguards Research and Development Association (1978).

8. L. KOCH and K. KAMMERICHS, "The Potential of Fission Product and Transplutonium Correlations in the Verification of the Reprocessing Input," *Proceedings of the 3rd Annual Symposium on Safeguards and Nuclear Material Management*, Karlsruhe, West Germany, May 6-8, p. 175, European Safeguards Research and Development Association (1981).

9. L. KOCH and S. SCHOOF, "The Isotope Correlation Experiment," EUR 776 EN, Commission of the European Communities (1983).

10. C. FOGGI, F. FRENQUELLUCCI, and G. PERDISA, "Isotope Correlations Based on Fission-Product Nuclides in LWR Irradiated Fuels," IAEA-SM-201/44, International Atomic Energy Agency (1976).

11. C. FOGGI, "Activities of the European Safeguards Research and Development Association (ESARDA) in the Field of Isotopic Correlations," IAEA-SM-231/26, International Atomic Energy Agency (1978).

12. C. FOGGI and C. PERDISA, "A Theoretical Analysis of the Isotopic Correlations Based on Stable Fission Products in LWR Irradiated Fuels," *Proceedings of the 1st Annual Symposium on Safeguards and Nuclear Material Management,* Vienna, October 2-6, p. 197, European Safeguards Research and Development Association (1979).

13. M. OHKUBO, "Gaseous Isotope Correlation Technique for Safeguards at Reprocessing Facilities," IAEA-STR-240, International Atomic Energy Agency (1988).

14. T. C. CHAPMAN, "Noble Gas, A Potential Nuclear Proliferation Indicator," *Trans. Am. Nucl. Soc.*, 68, p. 70 (1993).

15. G. B. HUDSON, "Noble Gas Isotope Measurements for Spent Nuclear Fuel Reprocessing: IAEA Task 90/0A211 Interim Report," UCRL-ID-118658, Lawrence Livermore National Laboratory (1993).

16. Y. AREGBE, S. VALKIERS, K. MAYER, and P. DE BIEVRE, "Comparative Isotopic Measurements on Xenon and Krypton," *Int. J. Mass Spec. Ion Processes*, 153, p. L1 (1996).

17. Y. AREGBE, K. MAYER, S. VALKIERS, and P. DE BIEVRE, "Release of Anthropogenic Xenon to the Atmosphere: a Large-Scale Isotope Dilution," *Int. J. Mass Spec. Ion Processes*, 154, p. 89 (1996).

18. Y. AREGBE, K. MAYER, S. VALKIERS, and P. DE BIEVRE, "Detection of Reprocessing Activities Through Stable Isotope Measurements of Atmospheric Noble Gas," *Fresnius J. Anal. Chem.*, 358, p. 533 (1997).

19. Y. AREGBE, "Meteorological Certification Measurements of Stable Isotope Amount Ratios of Xenon and Krypton," Ph.D. Dissertation, University of Technology in Vienna, Technical Sciences (1998).

20. U. FISCHER and H. W. WIESE, "Improved and Consistent Determination of the Nuclear Inventory of Spent PWR Fuel on the Basis of Cell Burnup Methods Using KORIGEN," KfK 3014, Karlsruhe Nuclear Research Center (1983).

21. C. W. NAKHLEH, R. T. PERRY, J. POTHS, W. D. STANBRO, W. B. WILSON, and B. L. FEAREY, "Noble Gas Atmospheric Monitoring at Reprocessing Facilities," presented at the International Workshop on the Status of Measurement Techniques for the Identification of Nuclear Signatures, Geel, Belgium, February 23-27, 1997.

22. C. W. NAKHLEH, W. D. STANBRO, L. N. HAND, R. T. PERRY, W. B. WILSON, and B. L. FEAREY, "Noble-Gas Atmospheric Monitoring for International Safeguards at Reprocessing Facilities," *Science and Global Security*, 6, p. 357 (1997).

23. W.S.CHARLTON, "Noble Gas Measurement and Analysis Technique for Monitoring Reprocessing Facilities," LA-13636-T, Los Alamos National Laboratory (1999).

24. E.A.VILLARINO, R.J.J.STAMM'LER, A.A.FERRI, J.J.Casal, "HELIOS: Angularly Dependent Collision Probabilities," *Nuclear Science and Engineering*, Vol.112, No.1, p.16 (1992).

25. D.I.POSTON, H.R.TRELLUE, "User's Manual, Version 1.00, for Monteburns, Version 3.01," LA-UR-98-2718, Los Alamos National Laboratory (1998).

国際単位系（SI）

表１．SI 基本単位

基本量	SI 基本単位	
	名称	記号
長さ	メートル	m
質量	キログラム	kg
時間	秒	s
電流	アンペア	A
熱力学温度	ケルビン	K
物質量	モル	mol
光度	カンデラ	cd

表２．基本単位を用いて表されるSI組立単位の例

組立量	SI 基本単位	
	名称	記号
面積	平方メートル	m^2
体積	立法メートル	m^3
速さ，速度	メートル毎秒	m/s
加速度	メートル毎秒毎秒	m/s^2
波数	毎メートル	m-1
密度（質量密度）	キログラム毎立法メートル	kg/m^3
質量体積（比体積）	立法メートル毎キログラム	m^3/kg
電流密度	アンペア毎平方メートル	A/m^2
磁界の強さ	アンペア毎メートル	A/m
（物質量の）濃度	モル毎立方メートル	mol/m^3
輝度	カンデラ毎平方メートル	cd/m^2
屈折率	（数 の） 1	1

表５．SI 接頭語

乗数	接頭語	記号	乗数	接頭語	記号
10^{24}	ヨタ	Y	10^{-1}	デシ	d
10^{21}	ゼタ	Z	10^{-2}	センチ	c
10^{18}	エクサ	E	10^{-3}	ミリ	m
10^{15}	ペタ	P	10^{-6}	マイクロ	µ
10^{12}	テラ	T	10^{-9}	ナノ	n
10^{9}	ギガ	G	10^{-12}	ピコ	p
10^{6}	メガ	M	10^{-15}	フェムト	f
10^{3}	キロ	k	10^{-18}	アト	a
10^{2}	ヘクト	h	10^{-21}	ゼプト	z
10^{1}	デカ	da	10^{-24}	ヨクト	y

表３．固有の名称とその独自の記号で表されるSI組立単位

組立量	SI 組立単位			
	名称	記号	他のSI単位による表し方	SI基本単位による表し方
平面角	ラジアン[(a)]	rad		$m\cdot m^{-1}=1$[(b)]
立体角	ステラジアン[(a)]	sr[(c)]		$m^2\cdot m^{-2}=1$[(b)]
周波数	ヘルツ	Hz		s^{-1}
力	ニュートン	N		$m\cdot kg\cdot s^{-2}$
圧力，応力	パスカル	Pa	N/m^2	$m^{-1}\cdot kg\cdot s^{-2}$
エネルギー，仕事，熱量	ジュール	J	$N\cdot m$	$m^2\cdot kg\cdot s^{-2}$
工率，放射束	ワット	W	J/s	$m^2\cdot kg\cdot s^{-3}$
電荷，電気量	クーロン	C		$s\cdot A$
電位差（電圧），起電力	ボルト	V	W/A	$m^2\cdot kg\cdot s^{-3}\cdot A^{-1}$
静電容量	ファラド	F	C/V	$m^{-2}\cdot kg^{-1}\cdot s^4\cdot A^2$
電気抵抗	オーム	Ω	V/A	$m^2\cdot kg\cdot s^{-3}\cdot A^{-2}$
コンダクタンス	ジーメンス	S	A/V	$m^{-2}\cdot kg^{-1}\cdot s^3\cdot A^2$
磁束	ウエーバ	Wb	$V\cdot s$	$m^2\cdot kg\cdot s^{-2}\cdot A^{-1}$
磁束密度	テスラ	T	Wb/m^2	$kg\cdot s^{-2}\cdot A^{-1}$
インダクタンス	ヘンリー	H	Wb/A	$m^2\cdot kg\cdot s^{-2}\cdot A^{-2}$
セルシウス温度	セルシウス度[(d)]	℃		K
光束	ルーメン	lm	$cd\cdot sr$[(c)]	$m^2\cdot m^{-2}\cdot cd=cd$
照度	ルクス	lx	lm/m^2	$m^2\cdot m^{-4}\cdot cd=m^{-2}\cdot cd$
（放射性核種の）放射能	ベクレル	Bq		s^{-1}
吸収線量，質量エネルギー分与，カーマ	グレイ	Gy	J/kg	$m^2\cdot s^{-2}$
線量当量，周辺線量当量，方向性線量当量，個人線量当量，組織線量当	シーベルト	Sv	J/kg	$m^2\cdot s^{-2}$

(a) ラジアン及びステラジアンの使用は、同じ次元であっても異なった性質をもった量を区別するときの組立単位の表し方として利点がある。組立単位を形作るときのいくつかの用例は表４に示されている。

(b) 実際には、使用する時には記号rad及びsrが用いられるが、習慣として組立単位としての記号“１”は明示されない。

(c) 測光学では、ステラジアンの名称と記号srを単位の表し方の中にそのまま維持している。

(d) この単位は、例としてミリセルシウス度m℃のようにSI接頭語を伴って用いても良い。

表４．単位の中に固有の名称とその独自の記号を含むSI組立単位の例

組立量	SI 組立単位		
	名称	記号	SI 基本単位による表し方
粘度	パスカル秒	$Pa\cdot s$	$m^{-1}\cdot kg\cdot s^{-1}$
力のモーメント	ニュートンメートル	$N\cdot m$	$m^2\cdot kg\cdot s^{-2}$
表面張力	ニュートン毎メートル	N/m	$kg\cdot s^{-2}$
角速度	ラジアン毎秒	rad/s	$m\cdot m^{-1}\cdot s^{-1}=s^{-1}$
角加速度	ラジアン毎平方秒	rad/s^2	$m\cdot m^{-1}\cdot s^{-2}=s^{-2}$
熱流密度，放射照度	ワット毎平方メートル	W/m^2	$kg\cdot s^{-3}$
熱容量，エントロピー	ジュール毎ケルビン	J/K	$m^2\cdot kg\cdot s^{-2}\cdot K^{-1}$
質量熱容量（比熱容量），質量エントロピー	ジュール毎キログラム毎ケルビン	$J/(kg\cdot K)$	$m^2\cdot s^{-2}\cdot K^{-1}$
質量エネルギー（比エネルギー）	ジュール毎キログラム	J/kg	$m^2\cdot s^{-2}\cdot K^{-1}$
熱伝導率	ワット毎メートル毎ケルビン	$W/(m\cdot K)$	$m\cdot kg\cdot s^{-3}\cdot K^{-1}$
体積エネルギー	ジュール毎立方メートル	J/m^3	$m^{-1}\cdot kg\cdot s^{-2}$
電界の強さ	ボルト毎メートル	V/m	$m\cdot kg\cdot s^{-3}\cdot A^{-1}$
体積電荷	クーロン毎立方メートル	C/m^3	$m^{-3}\cdot s\cdot A$
電気変位	クーロン毎平方メートル	C/m^2	$m^{-2}\cdot s\cdot A$
誘電率	ファラド毎メートル	F/m	$m^{-3}\cdot kg^{-1}\cdot s^4\cdot A^2$
透磁率	ヘンリー毎メートル	H/m	$m\cdot kg\cdot s^{-2}\cdot A^{-2}$
モルエネルギー	ジュール毎モル	J/mol	$m^2\cdot kg\cdot s^{-2}\cdot mol^{-1}$
モルエントロピー，モル熱容量	ジュール毎モル毎ケルビン	$J/(mol\cdot K)$	$m^2\cdot kg\cdot s^{-2}\cdot K^{-1}\cdot mol^{-1}$
照射線量（X線及びγ線）	クーロン毎キログラム	C/kg	$kg^{-1}\cdot s\cdot A$
吸収線量率	グレイ毎秒	Gy/s	$m^2\cdot s^{-3}$
放射強度	ワット毎ステラジアン	W/sr	$m^4\cdot m^{-2}\cdot kg\cdot s^{-3}=m^2\cdot kg\cdot s^{-3}$
放射輝度	ワット毎平方メートル毎ステラジアン	$W/(m^2\cdot sr)$	$m^2\cdot m^{-2}\cdot kg\cdot s^{-3}=kg\cdot s^{-3}$

表６．国際単位系と併用されるが国際単位系に属さない単位

名称	記号	SI 単位による値
分	min	1 min=60s
時	h	1h =60 min=3600 s
日	d	1 d=24 h=86400 s
度	°	1° =(π/180) rad
分	’	1’ =(1/60)° =(π/10800) rad
秒	”	1” =(1/60)’ =(π/648000) rad
リットル	l、 L	1l=1 $dm^3=10^{-3}m^3$
トン	t	1t=10^3 kg
ネーパ	Np	1Np=1
ベル	B	1B=(1/2) ln10(Np)

表７．国際単位系と併用されこれに属さない単位でSI単位で表される数値が実験的に得られるもの

名称	記号	SI 単位であらわされる数値
電子ボルト	eV	1eV=1.60217733(49)×10^{-19}J
統一原子質量単位	u	1u=1.6605402(10)×10^{-27}kg
天文単位	ua	1ua=1.49597870691(30)×10^{11}m

表８．国際単位系に属さないが国際単位系と併用されるその他の単位

名称	記号	SI 単位であらわされる数値
海里		1海里=1852m
ノット		1ノット=1海里毎時=(1852/3600)m/s
アール	a	1 a=1 $dam^2=10^2m^2$
ヘクタール	ha	1 ha=1 $hm^2=10^4m^2$
バール	bar	1 bar=0.1MPa=100kPa=1000hPa=10^5Pa
オングストローム	Å	1 Å=0.1nm=10^{-10}m
バーン	b	1 b=100$fm^2=10^{-28}m^2$

表９．固有の名称を含むCGS組立単位

名称	記号	SI 単位であらわされる数値
エルグ	erg	1 erg=10^{-7} J
ダイン	dyn	1 dyn=10^{-5}N
ポアズ	P	1 P=1 dyn・s/cm^2=0.1Pa・s
ストークス	St	1 St =1$cm^2/s=10^{-4}m^2/s$
ガウス	G	1 G ˆ10^{-4}T
エルステッド	Oe	1 Oe ˆ(1000/4π)A/m
マクスウェル	Mx	1 Mx ˆ10^{-8}Wb
スチルブ	sb	1 sb =1$cd/cm^2=10^4cd/m^2$
ホト	ph	1 ph=10^4lx
ガル	Gal	1 Gal =1$cm/s^2=10^{-2}m/s^2$

表10．国際単位に属さないその他の単位の例

名称	記号	SI 単位であらわされる数値
キュリー	Ci	1 Ci=3.7×10^{10}Bq
レントゲン	R	1 R = 2.58×10^{-4}C/kg
ラド	rad	1 rad=1cGy=10^{-2}Gy
レム	rem	1 rem=1 cSv=10^{-2}Sv
X線単位		1X unit=1.002×10^{-4}nm
ガンマ	γ	1γ=1 nT=10^{-9}T
ジャンスキー	Jy	1 Jy=$10^{-26}W\cdot m^{-2}\cdot Hz^{-1}$
フェルミ		1 fermi=1 fm=10^{-15}m
メートル系カラット		1 metric carat = 200 mg = 2×10^{-4}kg
トル	Torr	1 Torr = (101 325/760) Pa
標準大気圧	atm	1 atm = 101 325 Pa
カロリー	cal	
ミクロン	µ	1 µ =1µm=10^{-6}m

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