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Noble Gas Measurement and Analysis Technique for Monitoring Reprocessing Facilities

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NOBLE GAS MEASUREMENT AND ANALYSIS TECHNIQUE FOR MONITORING REPROCESSING FACILITIES.

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ABSTRACT

An environmental monitoring technique using analysis of stable noble gas isotopic ratios on-stack at a reprocessing facility was developed. This technique integrates existing technologies to strengthen safeguards at reprocessing facilities. The isotopic ratios are measured using a mass spectrometry system and are compared to a database of calculated isotopic ratios using a Bayesian data analysis method to determine specific fuel parameters (e.g., burnup, fuel type, fuel age, etc.). These inferred parameters can be used by investigators to verify operator declarations. A user-friendly software application (named NOVA) was developed for the application of this technique. NOVA included a Visual Basic user interface coupling a Bayesian data analysis procedure to a reactor physics database (calculated using the Monteburns 3.01 code system).

The integrated system (mass spectrometry, reactor modeling, and data analysis) was validated using on-stack measurements during the reprocessing of target fuel from a U.S. production reactor and gas samples from the processing of EBR-II fast breeder reactor driver fuel. These measurements led to an inferred burnup that matched the declared burnup with sufficient accuracy and consistency for most safeguards

applications. The NOVA code was also 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.

Work should continue to demonstrate the robustness of this system for production, power, and research reactor fuels.

I. INTRODUCTION

Preventing the spread of weapons of mass destruction (especially nuclear weapons) promotes the national security interests of the United States of America. Constructing a nuclear device requires the securing of weapons-useable nuclear material (either reprocessed plutonium or highly enriched uranium). Thus, detecting and curtailing the production of weapons-useable nuclear material is one way of impeding the proliferation of nuclear weapons. This nuclear material must be produced either through the enrichment of uranium (a process requiring large and relatively easy to detect facilities) or through the diversion of plutonium during the reprocessing of spent nuclear fuel.[1]

The International Atomic Energy Agency (IAEA) is generally charged with the safeguarding of nuclear materials at reprocessing facilities. The discovery in 1991 of a large, clandestine weapons program in Iraq led to a call for improvements in international safeguards on nuclear facilities and materials. The IAEA responded with an effort called "Programme 93+2". Programme 93+2 is intended to develop new and more efficient safeguards techniques.[2] The product of this effort is known as the "Model Protocol" and is described in the IAEA document INFCIRC/540.[3] INFCIRC/540 provides the basis for States to reach agreements with the IAEA on additional safeguards. Among the possible additions to safeguards contemplated in INFCIRC/540 is the use of environmental monitoring.

This dissertation follows the style and format of *Nuclear Technology*.

One particular challenge to IAEA safeguards is the monitoring of large scale reprocessing facilities devoted to civil uses.[4] Several such facilities have either recently become active or are scheduled to do so in the near future. In 1990, the UP-3 plant at La Hague, France initiated operations. The plant has a designed annual throughput of 800 tons of heavy metal.[5] In 1993, the Thermal Oxide Reprocessing Plant (THORP) in Great Britain began operation and is increasing its productivity to its designed throughput of 1200 tons of uranium per year.[6] The Tokai Reprocessing Plant (TRP) in Japan is currently operating with a throughput of 90 tons of heavy metal per year,[7] and the Rokkasho Reprocessing Plant (RRP) in Japan is now under construction with operations scheduled to start in the year 2000. RRP is designed for an annual throughput of 800 tons of uranium.[7] Safeguarding these facilities and similar plants in the future will pose an interesting challenge to the IAEA safeguards system. One environmental monitoring (EM) technique that may prove widely applicable to the safeguarding of reprocessing plants is the monitoring of noble gases contained in the facility's stack gases.

A. Project Objectives

The objective of this work was to develop an environmental monitoring technique to allow for accurate and noninvasive monitoring of reprocessing facilities to determine if an increased proliferation hazard exists due to activities within those facilities. The monitoring will be achieved by analyzing stack gases. Development of the method will involve: (1) establishment of a measurement technique for determining noble gas concentrations in stack gases (above those due to natural

sources), (2) generation of a database of noble gas emissions characteristic of different spent fuel conditions, and (3) development of an analysis procedure for determining spent fuel attributes from specific measurements using the aforementioned database. The methods developed here will provide monitoring techniques capable of accurate and noninvasive detection of potential proliferation activities during spent fuel reprocessing. In addition, these techniques will contribute to the international safeguards efforts for quantifying and securing fissile material separated from spent fuel.

B. Theory and Background Information

Proper development and implementation of an environmental monitoring technique of the type proposed here requires thorough knowledge of the nuclear fuel cycle including possible points for the diversion of weapons-grade nuclear material. It is also necessary to understand the physics of fission product generation and depletion in a nuclear system.

1. The Nuclear Fuel Cycle and Routes to Nuclear Weapons

The nuclear fuel cycle (Fig. 1) comprises all operations from the mining of uranium ore to the final disposal of waste products. The "front-end" of the nuclear fuel cycle refers to the preparation of uranium for use in reactors. The "back-end" of the nuclear fuel cycle refers to operations performed on spent fuel (e.g., reprocessing and disposal).[8]

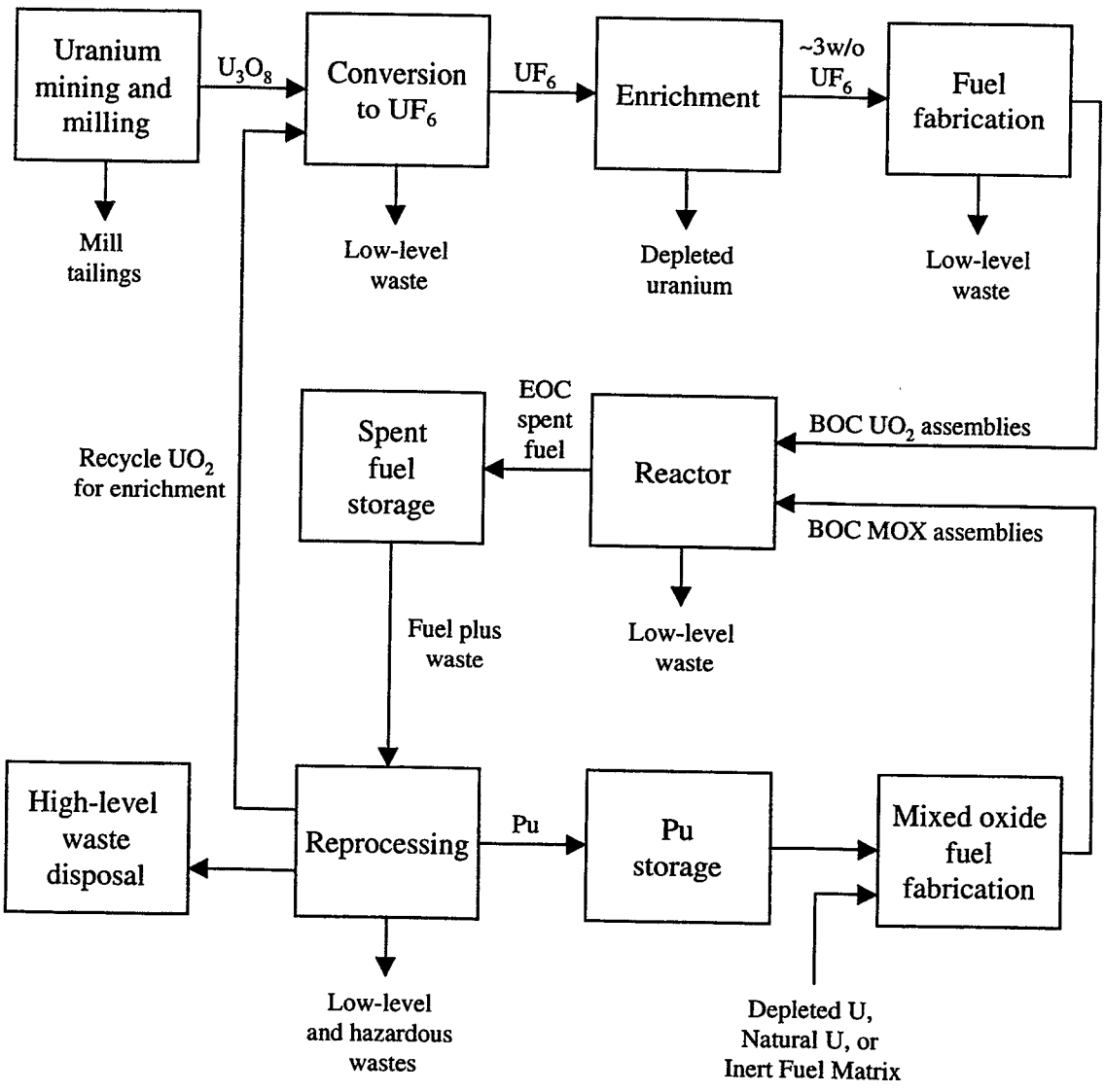

Fig. 1. The nuclear fuel cycle with plutonium and uranium recycle.

The first step in the nuclear fuel cycle is the mining of uranium ore. This ore is then reduced to the oxide U_3O_8, also called "yellow cake", through milling. This milling process leaves a large waste residue called "mill tailings". The yellow cake is

then converted to uranium hexafluoride (UF_6), a gas at ordinary temperatures and pressures. The UF_6 is then enriched in ^{235}U through a process called gaseous diffusion.[9]

Enrichment is the first point in the nuclear fuel cycle with the possibility of producing weapons-grade nuclear material. Most nuclear fuel is only slightly enriched (~3% w/o) and unsuitable for use in nuclear weapons; however, if the fuel was enriched to a high ^{235}U content (>20% w/o), it could potentially be used in the construction of a nuclear explosive. Enriching uranium requires large facilities and significant operational resources. Since these procedures are difficult to hide from inspectors, the proliferation hazards due to the enrichment process will be left for others to study.

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 uranium dioxide (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.[10] 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 the uranium 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 pressurized water reactor (PWR) fuel with a burnup of 33,000 MWd/MTU contains about 96% uranium, 1% plutonium, and 3% fission products.[10] The majority of the plutonium 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 cooling ponds. The first option is permanent disposal, most likely in a geological depository.[10] The second option is to attempt to recover the useable material from the spent fuel (i.e., the plutonium and uranium). Reprocessing refers to the procedure used to recover the plutonium and unburned uranium 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 in that it allows for the generation of separated plutonium, which can be used to produce nuclear explosives. Though it involves handling highly radioactive material, reprocessing of fuel only requires modest facilities and procedures. These facilities can easily be hidden or misused. For this reason, reprocessing is considered the most likely proliferation point in the nuclear fuel cycle and the primary thrust of the work presented here.

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 (uranium and plutonium) 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;[11] thus, the PUREX process will be described in more detail in this report.

The PUREX process (Fig. 2) consists of three cycles of solvent extraction using tri-n-butyl phosphate (TBP). PUREX uses liquid-liquid extraction principles and oxidation-reduction chemical reactions. 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. The oxides (fuel and fission products) go into solution, and the clad is left behind. This is the procedure for stainless steel or Zircaloy clad fuel; however, for aluminum clad fuel, the clad is dissolved using an aqueous solution of sodium hydroxide and sodium nitrate.[12] During the chopping and dissolving phases, gases (such as tritium, krypton, xenon, iodine, CO_2, nitrogen oxides, 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).[11]

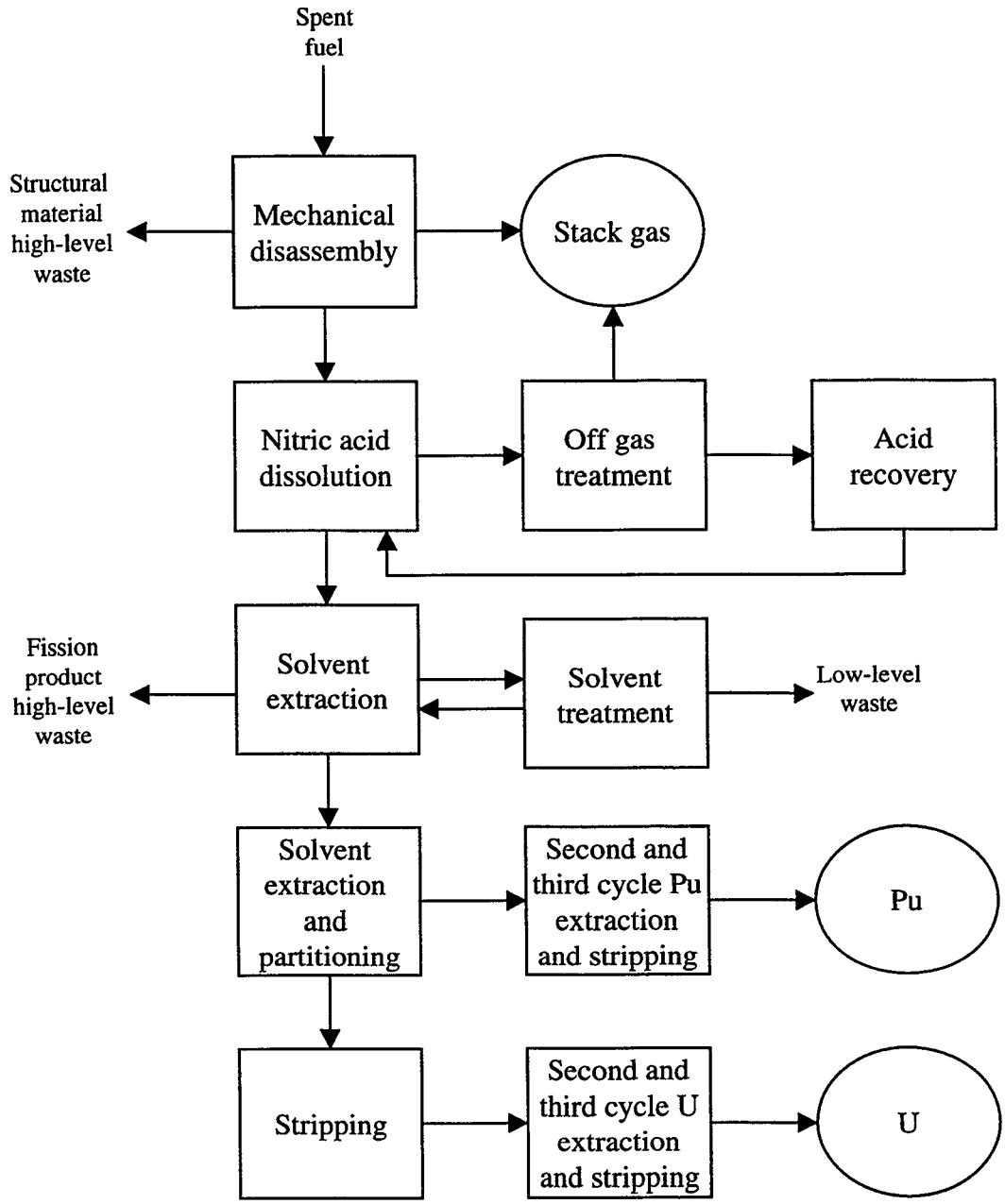

Fig. 2. The PUREX reprocessing process.

After dissolving, the nitric acid solution (containing uranium and plutonium) is processed through a solvent extraction that separates fission products and higher actinides from the uranium and plutonium. The uranium and plutonium are then separated using a chemical that reduces plutonium to an organic, insoluble state but not the uranium. This procedure makes use of the changing chemical properties of plutonium at higher oxidation states. For a complete discussion of reprocessing operations, see ref. 12.

The fission product gases released during the chop and dissolve phases include noble gases such as xenon and krypton. Due to their chemically inert nature, the xenon and krypton 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 most likely 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 due to their complex chemical and radioactive nature and their invasive sampling requirements, these are less attractive than the stack noble gases.

2. 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. Since 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.[9]

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.[9] As can be seen from Figs. 3 and 4 (with fission yields from ENDF/B-VI), fission products generally cluster around two peaks with respect to mass number. The xenon and krypton isotopes tend to be located on or near these peaks; however, the fission yields for krypton isotopes are usually between 5 to 10 times lower than those for the xenon isotopes. Figures 3 and 4 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.

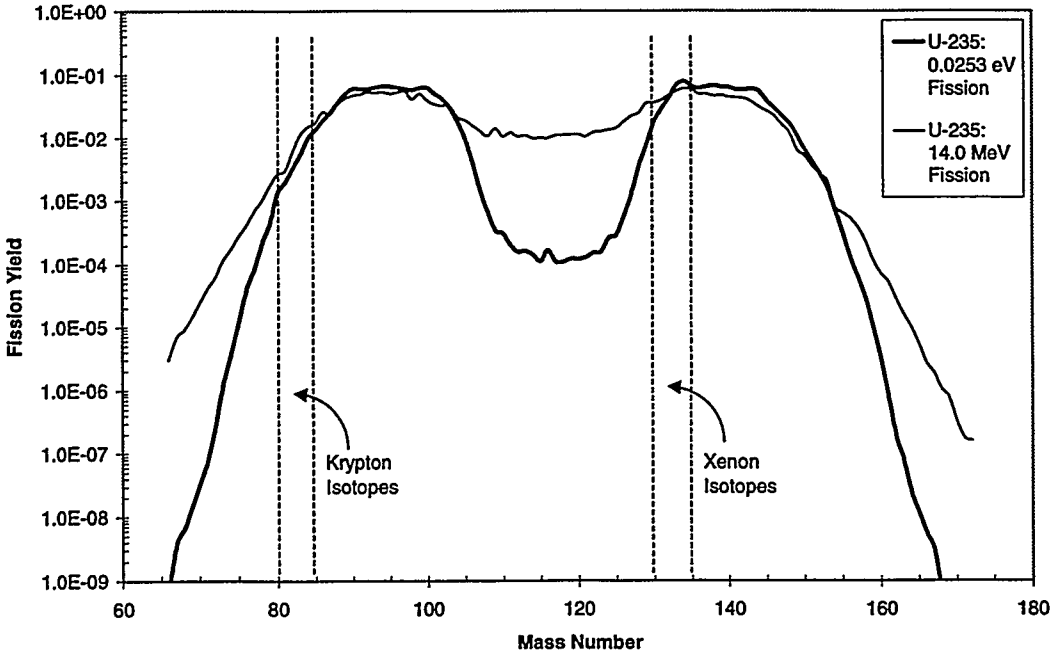

Fig. 3. ^{235}U thermal and 14 MeV fission yields versus mass number.

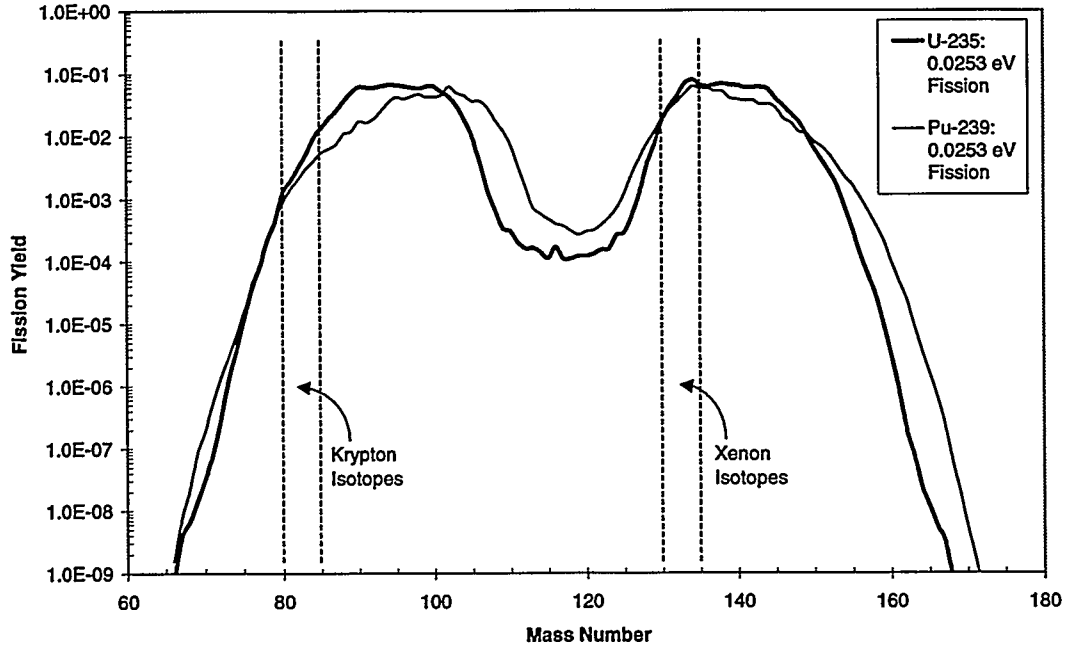

Fig. 4. ^{235}U and ^{239}Pu thermal fission yields versus mass number.

3. 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 krypton) are prevalent in spent fuel (see Figs. 3 and 4), yielding a large potential signal for measurement. Also, the relative concentrations of the xenon and krypton 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 uranium and plutonium from spent fuel. In addition, they are difficult and costly to remove from the stack gases. Thus, they are usually released directly into the lower atmosphere through the stack.

Another characteristic of the xenon and krypton isotopes that prove attractive for safeguards purposes is their low concentration in natural air. Generally speaking, background air contains approximately 87 ppb xenon and 1140 ppb krypton.[13] This implies that even low signatures of xenon and krypton released from a facility could be detected with a reasonably high degree of accuracy. Figures 5 and 6 contain plots of the percent isotopic compositions of the xenon and krypton isotopes in natural air[14] 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 xenon and krypton isotopes in natural air is markedly different from that produced in the gaseous elements from 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, operation 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 which is not produced during fission (see Section V.A.). Below is an examination of the xenon and krypton 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.

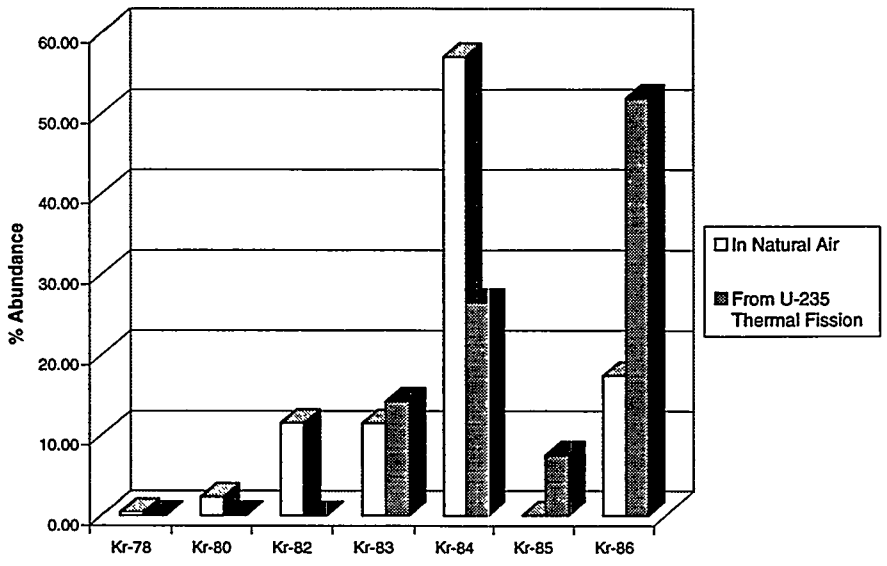

Fig. 5. Percent isotopic abundance of various krypton isotopes in natural air and from ^{235}U thermal fission.

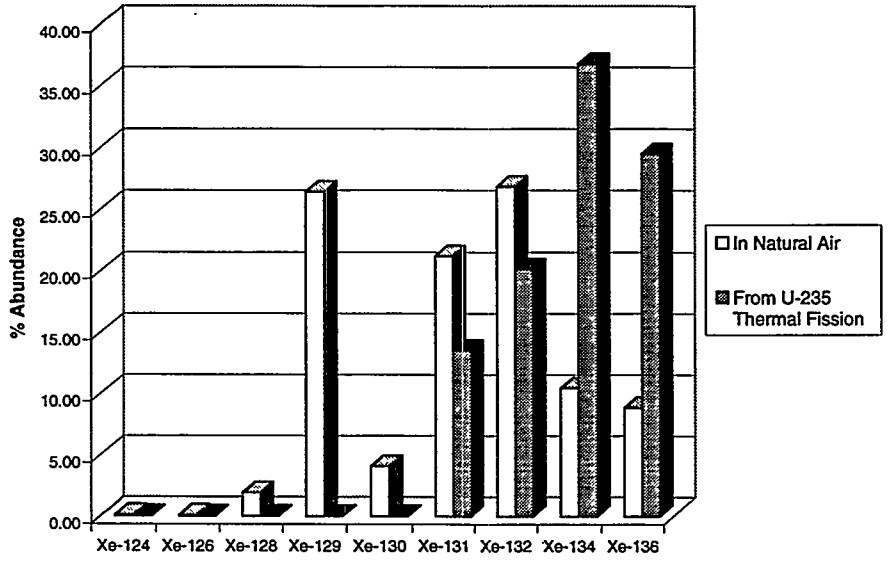

Fig. 6. Percent isotopic abundance of various xenon isotopes in natural air and from ^{235}U thermal fission.

Figures 7 and 8 illustrate the production and destruction modes (fission, absorption, and decay) for the krypton and xenon 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 the stable xenon and krypton isotopes are then produced by the β^- decay of parent isotopes. The direct fission yields for the stable isotopes are fairly small. Thus, in some cases there exists an appreciable time delay from fission to the generation of the stable xenon and krypton 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 xenon and krypton isotopes.

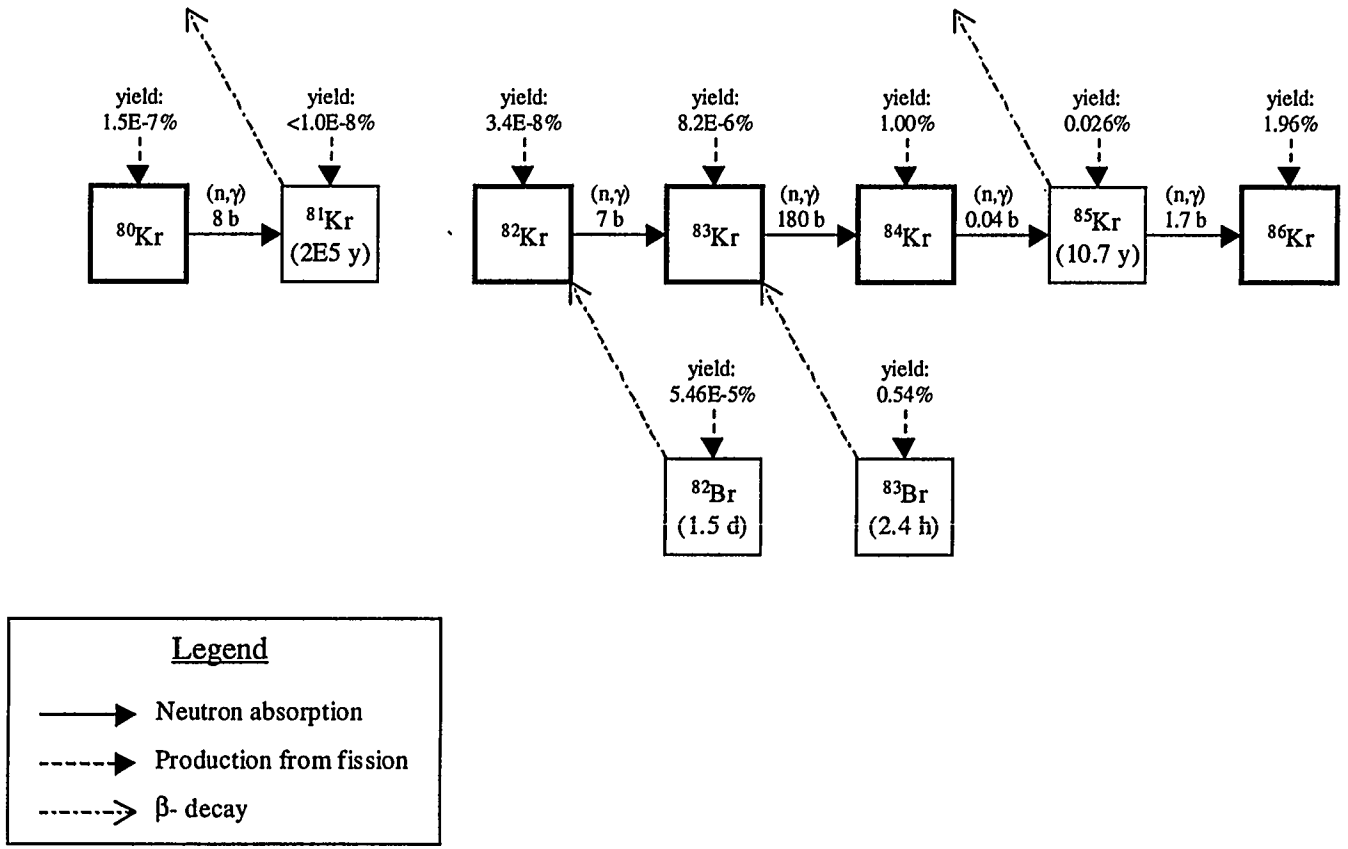

Fig. 7. Krypton isotope production and decay chains (information from ENDF/B-VI and ref. 15).

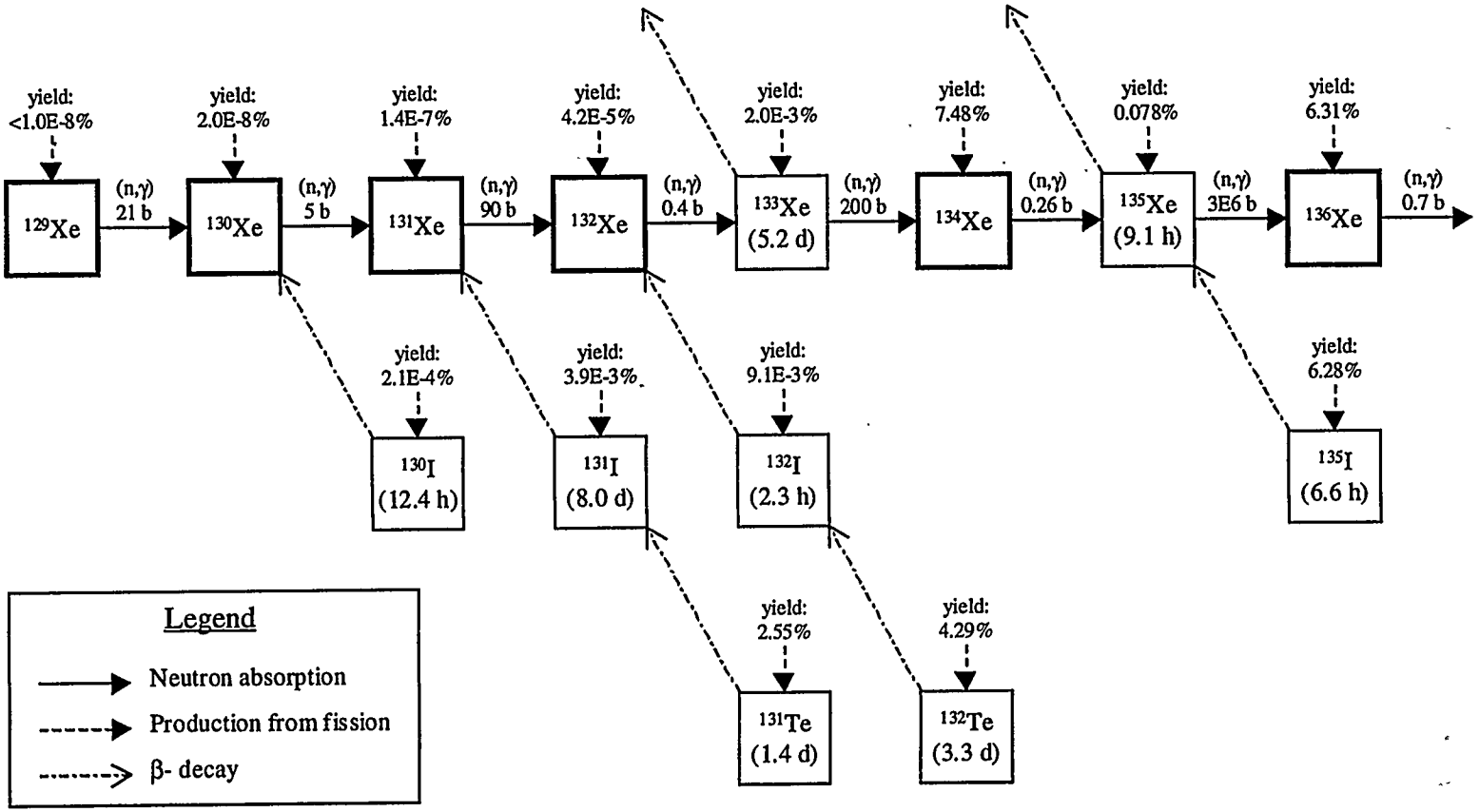

Fig. 8. Xenon isotope production and decay chains (information from ENDF/B-VI and ref. 15).

^{80}Kr and ^{81}Kr 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 krypton 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 (see Section V.A.). ^{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 krypton 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 (see Section V.A.).

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 Fig. 5).

^{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 the krypton 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. 5). 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. 6 and 8 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 b); 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 (see Section V.A.).

^{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 the xenon 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 xenon 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 b); 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 b), 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.

4. Low-Burnup Versus High-Burnup Fuel

One of the primary objectives of this study is to design a monitoring system to allow the 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 the plutonium 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 the ^{239}Pu is converted into the higher mass plutonium isotopes (especially the non-fissile ^{240}Pu and ^{242}Pu). These higher mass plutonium isotopes tend to be neutron poisons and make the potentially separated plutonium less attractive as a weapons material.

Generally speaking, one of the most notable parameters in determining the quality of the separated plutonium is the $^{240}Pu/^{239}Pu$ isotopic ratio. Figure 9 shows the variation of the $^{240}Pu/^{239}Pu$ isotopic ratio as a function of burnup for a U.S. PWR. Weapons-grade plutonium is usually considered to have a $^{240}Pu/^{239}Pu$ isotopic ratio around 7%.[16] As can be seen in Fig. 9, the $^{240}Pu/^{239}Pu$ isotopic ratio increases sharply with burnup and approaches a value of between 50 to 80% at high burnups.

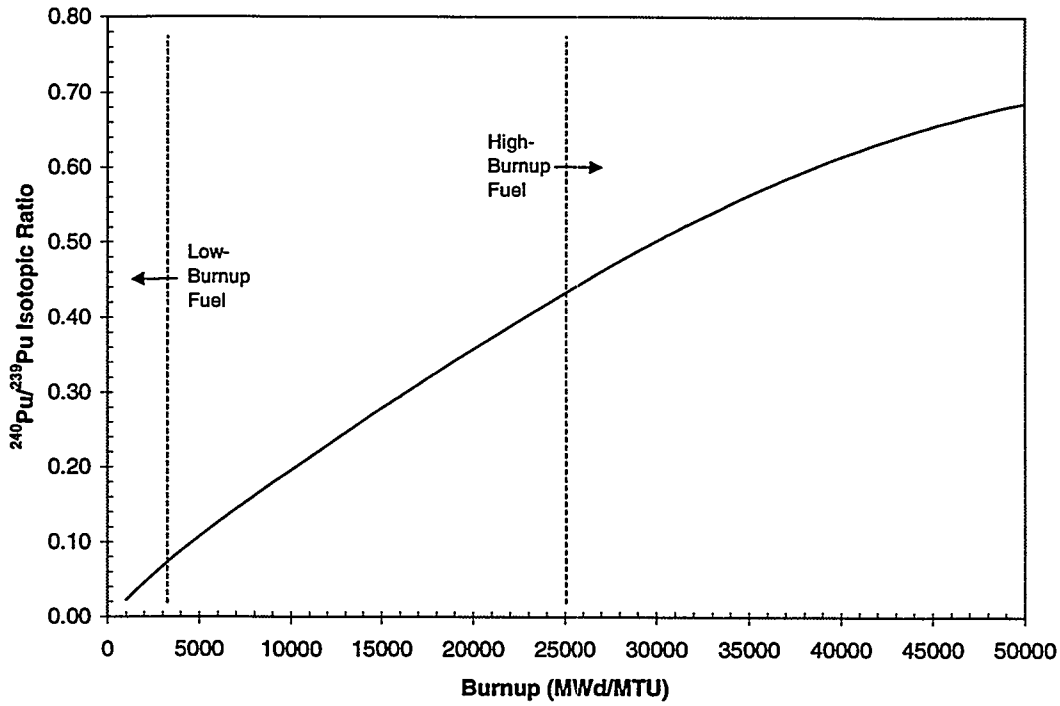

Fig. 9. $^{240}Pu/^{239}Pu$ isotopic ratio versus burnup for a U.S. PWR calculated using the HELIOS[17] lattice physics code.

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):

1. Distinguish low burnup from high burnup fuels,
2. Determine the spent fuel burnup,
3. Determine the reactor type which produced the fuel,
4. Determine other fuel parameters (including age, history, etc.),
5. Determine the $^{240}Pu/^{239}Pu$ isotopic ratio of the fuel,
6. Determine the plutonium content of the fuel.

With these characteristics an inspector would have the capability to confirm the information declared by the reprocessing facility and to determine if a proliferation hazard exists due to activities within the facility.

C. 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 the same element.[18] 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.[19-23] 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 cesium and neodymium isotopes in addition to the xenon and krypton isotopes. The xenon and krypton 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.[24-26] Again their work was heavily focused on heavy metal and radioactive fission products, but some effort analyzed the use of stable xenon and krypton 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 xenon and krypton gas for enhancing safeguards at reprocessing facilities.[27] 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.[28] The studies were preliminary in nature. One conclusion of these studies was that the xenon fission isotopes were easier to measure and contained more information than the krypton isotopes. They proposed a measurement technique which included separation of the xenon 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 xenon and krypton fission gases for monitoring reprocessing activities.[29] This study concentrated on determining the burnup and plutonium 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 xenon and krypton 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 xenon and krypton gases at reprocessing facilities.[30-33] This study made significant improvements in measurement techniques applied and treated in detail the atmospheric dilution problem. Aregbe used the KORIGEN code[34] 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 xenon and krypton noble gases for use in monitoring reprocessing facilities.[35-36] These studies were mostly exploratory in nature and suffered from the use of overly simplified reactor models. However, one major improvement in the 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, to date there does not exist 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. The objective of this work is to produce such a system and to validate it for use with several fuel types.

II. MONITORING SYSTEM OVERVIEW

The verification technique developed in this project couples a precise noble gas measurement system with an accurate fissiogenic gas database through the use of a sophisticated data analysis technique. The complete integrated system is illustrated in Fig. 10. 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 on-stack at a reprocessing facility during the chopping and/or dissolution of spent fuel. The gas samples are then analyzed in a system capable of determining the xenon and krypton isotopic ratios in the sample. The measurement system requires high-precision, fast sample processing, and the ability to aid in the removal of background air.

The reactor physics database contains xenon and krypton fissiogenic isotopic ratios and plutonium concentrations as a function of burnup for an exhaustive set of fuel types. These ratios and plutonium 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 plutonium, xenon, and krypton 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 xenon, krypton, and plutonium in as many reactor types as data allowed.

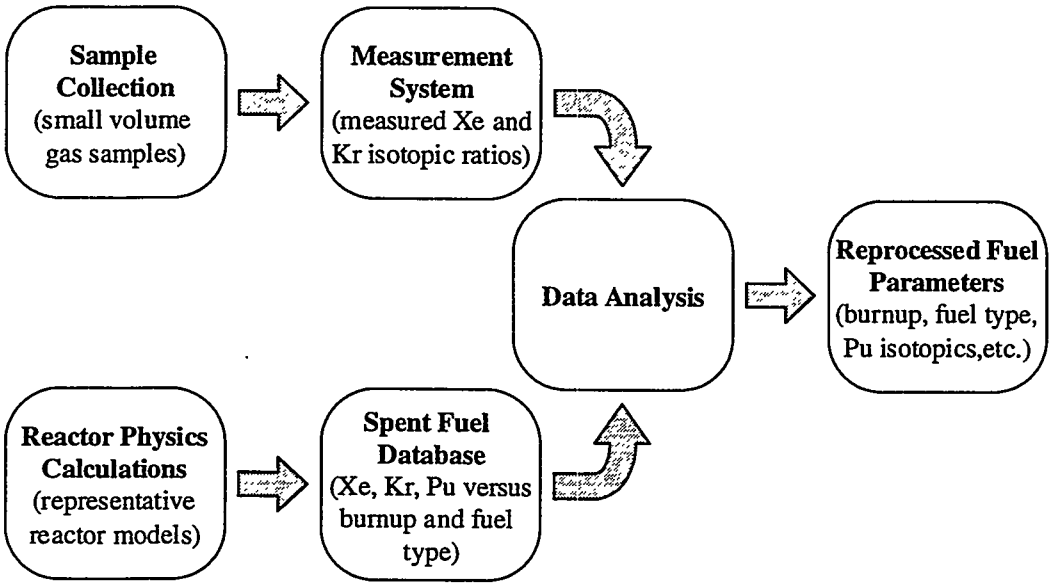

Fig. 10. Verification technique overview.

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, boiling water reactor (BWR), Canada Deuterium-Uranium (CANDU), etc.], fuel burnup, and plutonium 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 on-stack at 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 on-stack 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 plutonium composition) was generally less accurate than that from the Forward Problem; however, the Inverse Problem did not require information from the operator.

Also, the resulting confidences reported in these two problems were fundamentally different. The resulting confidence for the Inverse Problem was representative of the probability that the fuel being reprocessed is of a specific type at a certain burnup assuming that the database contains an exhaustive, mutually exclusive set of fuel types. The resulting confidence for the Forward Problem simply represented the probability that an inspector would measure the set of noble gas isotopic ratios given the fuel reprocessed is of the selected type.

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 plutonium concentrations in the fuel, then the Forward Problem allows the inspector the ability to accurately predict plutonium 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 plutonium isotopics using measured xenon and/or krypton 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.

III. MEASUREMENT SYSTEM

One of the most important parts of the verification method was the measurement system. Determining spent fuel parameters from noble gas emissions requires highly precise and accurate measurements of the isotopic ratios of the stable fission gases. Compared to the background air, the concentration of the fission gases on-stack at a reprocessing facility may be low. This necessitates the use of a high-precision instrument that will allow for the extraction of the fissiogenic component with a high-degree of accuracy. Since the isotopes of interest in this study are primarily stable noble gases, mass spectrometry is the ideal choice for a measurement system. Also note that all measurements are for isotopic ratios instead of absolute values. This allows for the neglecting of certain instrument characteristics and for greater generalization in the fissiogenic gas database.

A. Sample Collection

This technique makes use of gas samples taken on-stack at reprocessing facilities, ordinarily via isostatic sampling. The measurement system typically requires liter-sized gas samples that can be acquired using standard evacuated bottles. Generally, the dilution of the radioisotopes in the stack gases results in samples that present no radiological hazard and can be handled and transported without any special procedures. It may be possible to make use of the existing air-handling system at a given reprocessing facility to increase the ease with which these samples can be taken. The exact sampling protocol will likely be determined on a case-by-case basis. The

proposed technique allows for the direct removal of background air in the fissiogenic-gas-containing sample (see Section V.A. below); however, it is also recommended that background air samples be acquired to aid in confirmation of the removal of the natural air contaminant.

B. Mass Spectrometry System

There are two principal challenges associated with noble gas mass spectrometry for this project. First, the noble gas isotopic composition in stack gas samples must be analyzed with high accuracy to extract the fissiogenic noble gas signature in the presence of the more abundant atmospheric noble gases, which fortunately have different isotopic compositions. The second challenge is to develop robust sample analysis methods that will enable rapid, routine analysis of numerous stack gas samples.

These two goals are met through a new plasma-source, multi-collector mass spectrometer developed at LANL. This spectrometer currently provides xenon isotopic analysis on liter-sized air samples with a reproducibility of 0.05% in the isotopic ratios. Xenon was the initial focus in this study because of its generally larger fission yields and smaller background-air concentration. The krypton isotopes appear with a much higher concentration in natural air and have slightly smaller fission yields; thus, the measurement instrument's precision must be exceptional to acquire valuable information on fissiogenic krypton gases. The addition to the LANL instrument of detectors to determine krypton isotopic ratios is in the process of development. Though

the instrument is currently incapable of measuring krypton, the remainder of the verification project will include the possibility of krypton for use in monitoring reprocessing facilities in anticipation of the adaptation of the existing instrument.

An analysis on the LANL system takes less than 30 minutes and does not require concentration of xenon in the sample. Rapidity of analysis, simplicity of procedure, and modest required sample size are some of this system's major advantages compared to other state-of-the-art, dynamic, gas mass spectrometers (although the accuracy of this system maybe less).

This mass spectrometer utilizes the Mattauch-Herzog geometry,[37] with the xenon multi-collector located at a 24 cm radius in the extended focal plane of the magnet. High ionization efficiency is achieved by a 2.45 GHz microwave plasma ion source operated at low pressure (2.6×10^{-5} atm of argon).[38] Ions are accelerated through 8 kV from the ion source, pass through a quadrupole triplet lens system for beam shaping and into the instrument. Detection is through simultaneous collection of masses 129-136 with Faraday cups. To fit the tight spacing and tolerances (a 0.9 mm center-to-center spacing per mass unit and a slit width of 0.046 mm), the collectors are assembled as a stack of metalized ceramic plates with a nine-slit mask mounted in front. This collector assembly is located at the focal plane within the gap of the analyzed magnet, which provides excellent secondary electron suppression. Simultaneous detection of the xenon isotopes yields two primary advantages: (1) increased accuracy by compensating for fluctuations in ion beam intensity due to instability in the plasma source and (2) excellent duty cycle and thus sample utilization.

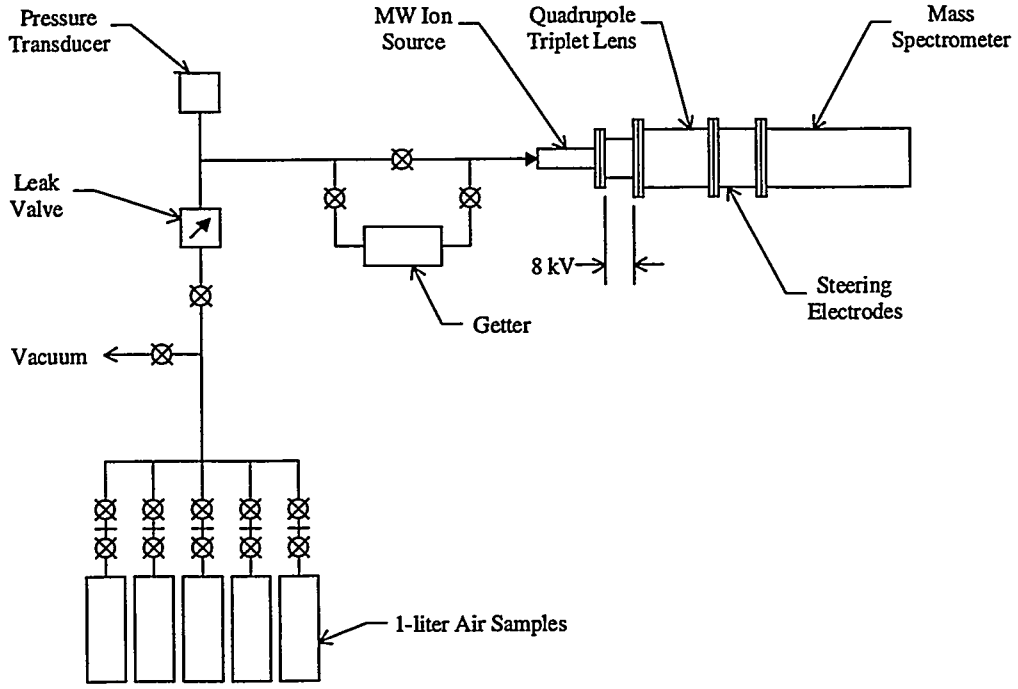

Fig. 11. Associated gas-handling system.

The associated gas-handling system (Fig. 11) and protocol to run the samples are simple by design. An air-like sample flows through a getter and into the ion source at a rate controlled by the leak valve and monitored on the pressure transducer. The getter removes all reactive gases, so that the sample enters the ion source as argon with traces of other noble gases (100 and 10 ppm of krypton and xenon, respectively). The argon naturally present in air provides the pressure needed to sustain the plasma. Once an ion beam is established with the sample, the peak shape and sensitivity are optimized with the quadrupole triplet lens system and steering electrodes. Data are

then collected for six stable, heavy xenon isotopes (^{129}Xe, ^{130}Xe, ^{131}Xe, ^{132}Xe, ^{134}Xe, and ^{136}Xe) and associated baselines over a period of time to monitor mass-bias effects and acquire adequate statistical data. Unknown sample analysis is alternated with xenon analysis on ambient air samples to provide data for the mass-bias correction and relative collector efficiencies. The fissiogenic component is then calculated based on the difference between the sample and the measured air values (see Section V.A.).

A further advantage of this system is minimal cross-talk between samples (or "memory effect"). No significant memory effect was detected when switching between atmospheric samples and fissiogenic samples that had up to a 50% overabundance of $^{136}Xe/^{132}Xe$ compared to the atmospheric value. Keeping the xenon in a matrix (air or argon) appears to help reduce sorption of xenon on the walls of the vacuum system.

The LANL instrument is a high-precision mass spectrometer and is available for the development of this technique; however, it may be determined after complete integration of the verification method that a simpler instrument (perhaps even a portable instrument) may be capable of being used at a reprocessing facility. The determination of the required measurement precision and accuracy will be determined from an analysis of the completed verification system.

IV. REACTOR PHYSICS CALCULATIONS

One of the oversights made by previous researchers studying this problem has been a marked lack of rigorous reactor physics calculations. An accurate methodology (including reactor models, reactor analysis codes, and data libraries) for calculating fissiogenic noble gas ratios is developed in this section. The first step in performing these calculations was to evaluate reactor analysis methodologies with sufficient accuracy for use in the on-stack environmental monitoring application. A series of available reactor physics codes (and data libraries) were examined to determine which was most capable of predicting the fissiogenic noble gas isotopic ratios for use in the database. A series of models for a set of representative reactor fuel types was then developed, and a database of fissiogenic noble gas isotopic ratios as a function of burnup and fuel type was produced for an exhaustive set of reactors.

A. Calculational Methodology

Studies have been performed using Monteburns 3.01 [39] to analyze the effects of various parameters on calculated assembly-level spent fuel isotopics for PWRs. Monteburns 3.01 is a newly developed code at LANL which links the Monte Carlo transport code MCNP[40] to the burnup and depletion code ORIGEN2.[41] These studies included determining effects of parameters such as the number of radial fuel regions used, burnup step sizes, and axial power profiles. The general motivation for this work was to create models for generating isotopic ratios with sufficient accuracy for use in the on-stack environmental monitoring technique; however the spent fuel isotopics are

also potentially useful for reprocessing calculations, shielding applications, and spent fuel disposal studies. Therefore, the aim was to construct a calculational scheme that can produce assembly-level isotopics for a wide-variety of reactor types with an emphasis on uses in correlating measured effluent characteristics with spent fuel burnup and perhaps plutonium compositions. Unless otherwise specified, for all of the Monteburns calculations included below, each MCNP case was run with 8000 particles per generation and 450 generations. The first 50 generations were excluded from the statistics, thus 3.2 MegaHistories (3.2 MH or 3.2 million active histories) were tallied per burnup time step.

1. Calculational Level

The first step in determining a formalism for calculating assembly-averaged spent fuel isotopics is to choose an appropriate calculational level. Five possible calculational levels can initially be considered: homogeneous (or point) calculation, pin cell, pin cell with assembly-level fuel-to-moderator ratio (called a modified pin cell), assembly-level, and full-core. The first calculational level (homogeneous calculation) is identical to that performed by ORIGEN2 alone. ORIGEN2 has been shown to be generally accurate to within ±2% for uranium and plutonium isotopics, ±10-20% for higher actinide isotopics, and ±10-15% for fission product isotopics.[42-44] Though these accuracies may be sufficient for some applications, better agreement is desired for the environmental monitoring system.

The pin cell level calculations consist of modeling a single pin cell from an assembly. This ignores the effects of water holes and burnable absorbers on the neutron flux spectrum. This level of calculation is relatively simple to generate and is not computationally intensive, even when considering axial effects.[45]

The third calculational level involves calculations using a modified pin cell. The modified pin cell utilizes the same fuel and cladding dimensions as the geometric pin cell but has a modified pin pitch. This modification is made by adjusting the modified pin cell's pitch so that the fuel-to-moderator volumetric (FM) ratio of the modified pin cell corresponds to the FM ratio of the assembly. For a square assembly in a PWR, this calculation is performed using the following equation:

$$P_{FM} = \frac{P_{asb}}{\sqrt{N_{pins}}} \tag{1}$$

where P_{FM} is the adjusted pin-to-pin pitch, P_{asb} is the assembly pitch, and N_{pins} is the number of pins per assembly. This conversion is illustrated in Fig. 12. Generally, this adjustment leads to a pin-to-pin pitch slightly larger than the unadjusted pitch.

Assembly-level calculations consist of explicitly modeling all pins within an assembly, as well as water holes, burnable absorbers, and any other structures. This level of calculation is computationally intensive for Monte Carlo codes but yields very accurate assembly-averaged results. It is this level of calculation that is used for determining spent fuel isotopics for most applications.[46]

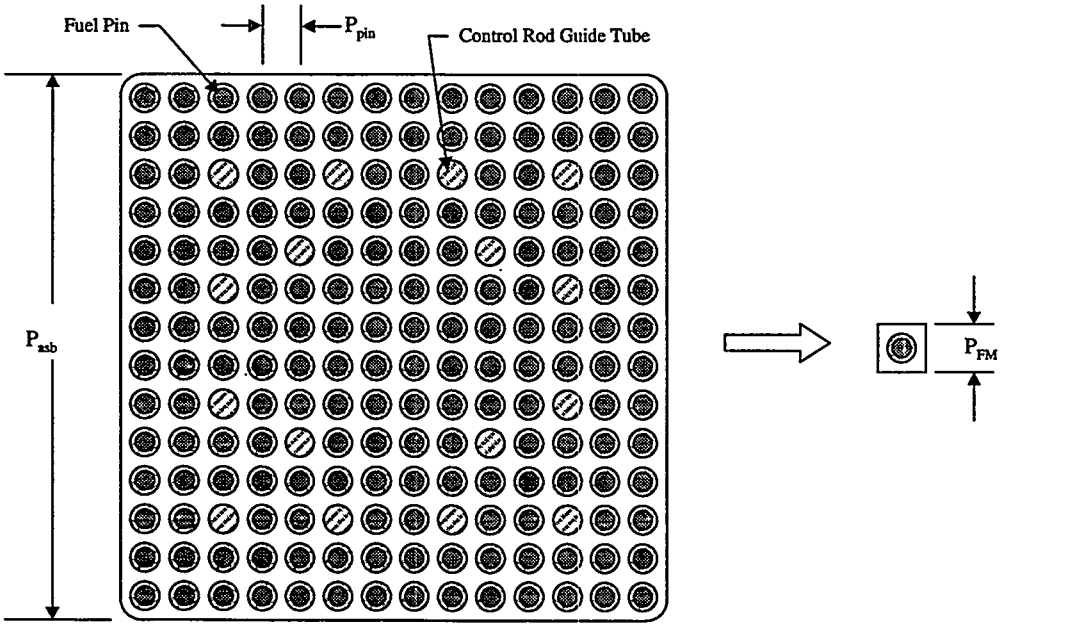

Fig. 12. Procedure for collapsing from assembly-level calculation to pin cell calculation.

Performing full-core calculations amount to adding the effects of radial core leakage and power peaking to the assembly-level calculations. These calculations require some knowledge of the core loading patterns (which will not be generally available to any inspector). Since, power and production reactors are neutronically large, the full-core effects are minimal on the spent fuel isotopics. Due to this and the much larger computational effort required, full-core calculations were eliminated from further consideration. Also it will be determined below that any full-core averaging procedure will be inherently accounted for in our model averaging.

TABLE I
Nominal PWR Assembly Parameters

Type	15 × 15 (square)
Assembly Pitch	21.402 cm
Modified Pin-to-Pin Pitch	1.4984 cm*
Fuel Pellet Diameter	0.932 cm
Clad Inner Diameter	0.948 cm
Clad Outer Diameter	1.072 cm
Fuel Density	10.00 g/cc
Fuel Enrichment	3.00 w/o ^{235}U
Active Fuel Length	365 cm
Clad Material	Zircaloy-4
Clad Density	6.56 g/cc
Coolant Material	light water
Coolant Density	0.7027 g/cc
Specific Power	30.0 W/g
Decay Time	150 days

* Adjusted to produce a pin cell with the same fuel-to-moderator ratio as an assembly.

To determine the best of the three levels (i.e., geometric pin cell, modified pin cell, and assembly-level) that yields isotopic data with sufficient accuracy for the gaseous effluent application, numerical experiments were performed with Monteburns based on typical assembly data for a model PWR assembly. The characteristics of this model PWR assembly are given in Table I. Calculations were performed for this assembly at the assembly-level, pin cell level, and modified pin cell level using a pin cell with the same FM ratio as the assembly.

Comparisons were made between each of the calculational levels for the following spent fuel isotopic ratios: $^{235}U/^{238}U$, $^{240}Pu/^{239}Pu$, $^{131}Xe/^{134}Xe$, $^{132}Xe/^{134}Xe$,

$^{136}Xe/^{134}Xe$, $^{83}Kr/^{86}Kr$, and $^{84}Kr/^{86}Kr$. The values of the individual isotopic ratios calculated were compared using the maximum percent deviation (MPD) of each result from a value calculated using some nominal parameter (either analytically determined, fine mesh limit, or arbitrarily chosen). The MPD is defined as follows:

$$MPD = \max_i \left[\frac{\left| x_i - x_i^{limit} \right|}{x_i^{limit}} \right] \cdot 100\% \qquad (2)$$

where x_i is a particular isotopic ratio at burnup step i and x_i^{limit} is the same isotopic ratio at burnup step i used as a reference value and calculated using an analytical, fine mesh, or nominal value of some parameter. If the nominal parameter being used is either an analytically determined result or the fine mesh limit result, then the value of the MPD represents the maximum relative difference in the isotopic ratio.

For the PWR assembly investigated, it was found that the modified pin cell yielded spent fuel isotopic ratios with MPDs relative to the assembly-level results of less than 0.038% for all ratios. The simple geometric pin cell yielded MPDs for all ratios from the assembly-level results of less than 0.851%. Thus, the modified pin cell was 22.4 times more accurate at reducing the maximum difference from the assembly-level results than the geometric pin cell. This implies that using a modified pin cell will generally result in isotopics representative of an assembly with a maximum isotopic ratio of less than 0.038%, which should be sufficient for the environmental monitoring technique proposed. Modified pin cell level calculations will therefore be used throughout the remainder of this document.

2. Number of Radial Fuel Regions

It is well known that within a given cylindrical fuel pin, the fission rate and neutron spectrum change from the surface of the pin to the center of the pin.[47] In fact, the central portions of a burned pin cell have relatively similar isotopic concentrations, whereas the thin surface layers of the pin can have radically different isotopic concentrations than the central region. For this reason, it is important to determine not only the appropriate number of radial, flat-flux fuel regions per pin for use in the burnup calculations, but also the distribution of those regions throughout the pin.

Numerical studies were performed using several fuel region distributions, including a linear separation of fuel regions, a quadratic separation of fuel regions, and an exponential separation of fuel regions. A linear distribution is the simplest way of determining the fuel region radii. The radii are given by

$$r(i) = R_{fo}\frac{i}{N_r} \qquad for \quad i = 1,2,3,\ldots,N_r \tag{3}$$

where $r(i)$ is the outer radius of fuel region i, R_{fo} is the fuel outer radius, and N_r is the total number of radial fuel regions. This distribution separates the radii evenly throughout the pin.

The quadratic distribution is essentially equivalent to setting all of the radial fuel region areas equal. This can be achieved using the following:

$$r(i) = R_{fo}\sqrt{\frac{i}{N_r}} \qquad for \quad i = 1,2,3,\ldots,N_r \tag{4}$$

where the variable definitions are the same as those given above.

The exponential distribution is suggested due to the attenuation of the low energy neutrons as they travel into the fuel. It was determined via numerical experimentation that an appropriate choice for the radial fuel region distribution in a cylindrical pin cell is given by

$$r(i) = R_{fo}\left[\frac{1-\exp(-\Sigma_a i)}{1-\exp(-\Sigma_a N_r)}\right] \qquad for \quad i = 1,2,3,\ldots,N_r \qquad (5)$$

where Σ_a is a one-group macroscopic absorption cross section and the other variables are the same as those mentioned above. For PWRs, the one-group cross section can be well approximated using the ^{235}U and ^{238}U thermal absorption cross section values. Thus, for 3.0 w/o ^{235}U fuel, $\Sigma_a \approx 0.55$ cm^{-1}. This value will change depending on the spectrum of neutrons in the reactor, the fuel composition, and the fuel density; however, for most LWR reactors, using a one-group cross section of 0.55 cm^{-1} yielded a fairly reasonable radial fuel region distribution. Figure 13 illustrates the effect of the exponential distribution on the radial fuel regions.

Numerical experiments were performed with Monteburns based on the PWR assembly specifications described in Table I with 1, 2, 3, 4, 5, 6, 7, 8, 9, and 12 fuel regions for each distribution method. The fine mesh limit was determined by taking the 12 fuel region case using the exponential distribution and adding the 12 regions from the linear distribution. It was found that this 24 region case yielded the same result (within five significant digits) as the 12 region case using the exponential distribution only. It was therefore assumed that the 12 region exponential distribution well approximates the fine mesh limit.

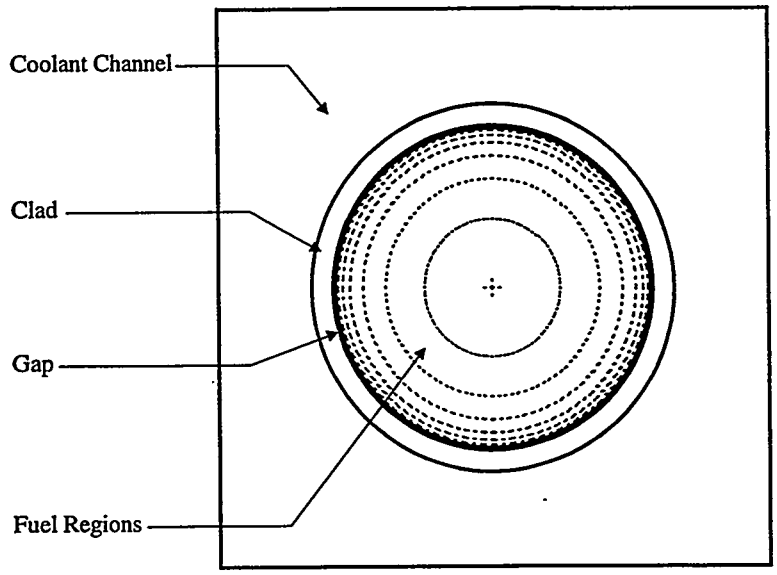

Fig. 13. Cross sectional view of calculational pin cell with seven exponentially determined radial fuel regions.

Comparisons were made for each of the seven isotopic ratios mentioned earlier. It was determined that using an appropriate exponential distribution for the radial fuel regions (with many more regions near the surface of the pin than the center of the pin) yielded a result indistinguishable from the fine mesh limit, with the fewest number of fuel regions, compared to the other proposed distributions. Figure 14 shows the convergence of each distribution method for the $^{131}Xe/^{134}Xe$ isotopic ratio. Similar results are found for each of the other isotopic ratios. In all cases, the MPDs for the exponential distribution decreased more rapidly than the MPDs for the other distributions.

The exponential fuel region distribution described in Eq. (5) was then used to determine the sensitivity of the eleven isotopic ratios to the number of fuel regions used. Numerical experiments were performed with Monteburns for the model PWR assembly described in Table I using 1, 2, 3, 4, 5, 6, 7, 8, 9, and 12 fuel regions. The spent fuel isotopic ratios calculated for these cases were then compared to the fine mesh limit results described earlier. The MPDs for these cases (calculated for burnups ranging from 0 to 50000 MWd/MTU with a 2000 MWd/MTU step size) are listed in Table II.

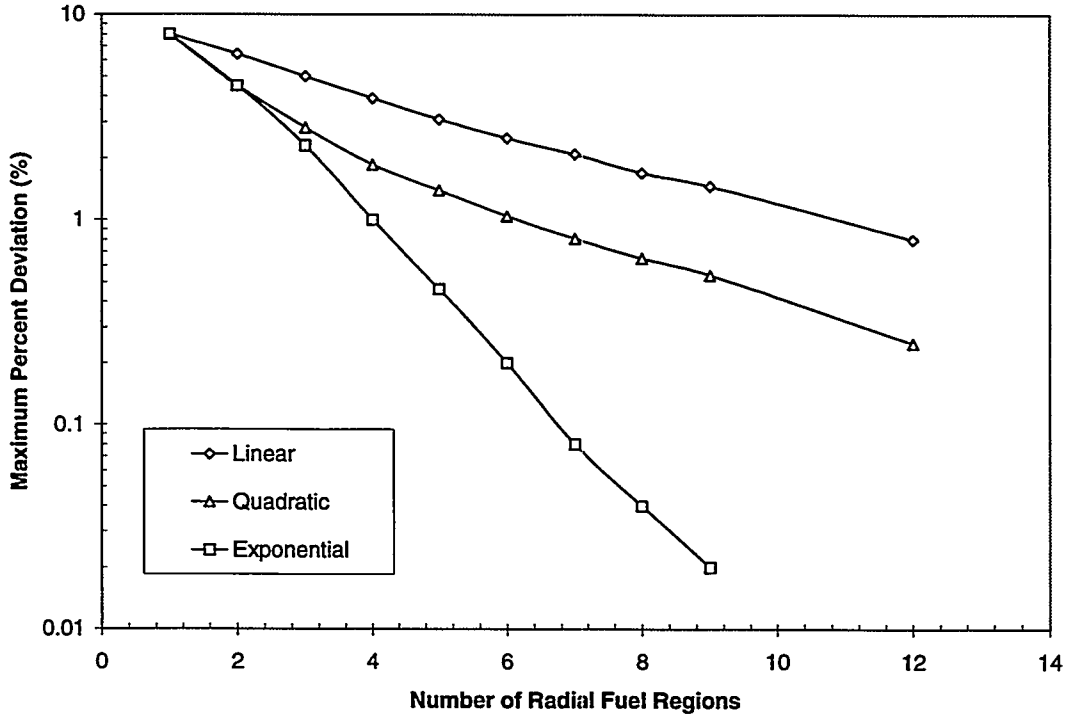

Fig. 14. Maximum percent deviation in the $^{131}Xe/^{134}Xe$ isotopic ratio from the fine mesh limit result as a function of the number of radial fuel regions used for three different distributions of the radii.

TABLE II
Maximum Percent Deviation of Various Isotopic Ratios from the Fine Mesh Value (Chosen at 21 Radial Fuel Regions) for Different Numbers of Flat-Flux Radial Fuel Regions

	Maximum Percent Deviation from Fine Mesh Limit (%)						
# of Fuel Regions	$^{235}U/^{238}U$	$^{240}Pu/^{239}Pu$	$^{83}Kr/^{86}Kr$	$^{84}Kr/^{86}Kr$	$^{131}Xe/^{134}Xe$	$^{132}Xe/^{134}Xe$	$^{136}Xe/^{134}Xe$
1	0.95	0.67	0.09	0.13	8.00	2.64	0.17
2	0.36	0.34	0.06	0.11	4.50	1.77	0.15
3	0.26	0.23	0.07	0.10	2.30	0.86	0.09
4	0.24	0.17	0.05	0.07	1.00	0.39	0.05
5	0.18	0.13	0.05	0.05	0.46	0.14	0.05
6	0.08	0.10	0.03	0.03	0.19	0.05	0.03
7	0.05	0.06	0.03	0.02	0.08	0.03	0.02
8	0.04	0.05	0.02	*	0.04	0.02	*
9	0.02	0.03	*	*	0.02	*	*
12	*	*	*	*	*	*	*

* Accurate to within five significant digits

As can be seen, the use of nine fuel regions yielded results within 0.03% of the fine mesh limit for all of the isotopic ratios. This degree of accuracy was deemed more than sufficient for the gaseous effluent monitoring application. Modified pin cells with nine flat-flux radial fuel regions were used to produce the isotopic ratios presented in the remainder of this dissertation.

3. Burnup Step Size

Generally, burnup codes make the assumption that the parameters influencing the depletion equations (i.e., one- or few-group cross sections, scalar fluxes, and fission

yields) do not change significantly during each burnup step. It is obvious that as the burnup step size is increased, this assumption will no longer be valid. To determine the appropriate burnup step size needed for accurately calculating the desired spent fuel isotopic ratios, the model PWR assembly from Table I was simulated in Monteburns for burnups from 0 to 50000 MWd/MTU using burnup step-sizes of 25000, 10000, 5000, 2000, 1000, 500, and 100 MWd/MTU. Spent fuel isotopic ratios from Monteburns were compared for all of these cases. It was determined that the 500 MWd/MTU case and the 100 MWd/MTU case agreed to within five significant digits. Thus, the 100 MWd/MTU case was taken to be a good approximant to the fine mesh limit.

The MPDs of the 25000, 10000, 5000, 2000, 1000, and 500 MWd/MTU cases as compared to the 100 MWd/MTU case (all calculated using nine flat-flux radial fuel regions for burnups ranging from 0 to 50000 MWd/MTU) are listed in Table III. As can be seen, the use of a burnup step size of 1000 MWd/MTU adds no significant uncertainty (<0.02%) to the calculated results for all of the isotopic ratios. In fact, the use of a burnup step size of 2000 MWd/MTU yields results within 0.04% of the fine mesh limit, which is probably sufficient for most applications including the proposed environmental monitoring technique. Modified pin cells with nine flat-flux radial fuel regions and a 2000 MWd/MTU burnup step size were used for the remainder of the calculations in this sensitivity study.

TABLE III
Maximum Percent Deviation of Various Isotopic Ratios from the Value at the Fine Mesh Limit (Chosen as 100 MWd/MTU) for Several Burnup Step Sizes (in MWd/MTU)

	Maximum Percent Deviation from Fine Mesh Limit (%)						
# of Fuel Regions	$^{235}U/^{238}U$	$^{240}Pu/^{239}Pu$	$^{83}Kr/^{86}Kr$	$^{84}Kr/^{86}Kr$	$^{131}Xe/^{134}Xe$	$^{132}Xe/^{134}Xe$	$^{136}Xe/^{134}Xe$
25,000	26.39	15.48	3.06	1.46	4.52	1.41	0.39
10,000	2.51	3.41	0.57	0.22	0.69	0.57	0.15
5,000	0.10	1.09	*	*	0.09	0.08	*
2,000	*	0.04	*	*	*	*	*
1,000	*	*	*	*	*	*	*
500	*	*	*	*	*	*	*

* Accurate to within five significant digits

4. Axial Profile Averaging

Axial isotopic distributions along a pin or assembly generally can be shown to be proportional to the time-integrated axial reactor power distribution.[48] The axial power distribution ordinarily takes a shape that is relatively flat along a central portion of the pin and then drops off rapidly near the ends. A general shape of this type was used to investigate the appropriate set of axial fuel regions for use in generating axially-averaged compositions for depletion studies. Note that axial effects such as voids in the coolant (crucial for boiling water reactors) and partial control rod insertion were not investigated in this study, the axial investigation here is purely to determine the most appropriate fuel regions for use in generating the pin cell axially-averaged (i.e., assembly) isotopic ratios mentioned earlier. Also note that since MCNP generates 3D

transport solutions to the neutron transport equation, the neutron flux shape in the axial direction will be accurate; however no supplementary power calculation was performed to generate appropriate temperature-dependent cross sections for each axial region. This effect will be studied in more detail later.

In all cases, symmetry about the center of the pin was assumed. Therefore, a reflecting boundary was used at $z=H/2$ (the center plane of the pin). Also, when it is stated that two axial fuel regions are being used, the use of the reflection boundary condition implies that two regions are present in the bottom of the core and two identical regions are present in the top of the core (for a total of four axial fuel regions). Also, note that the uniform axial case implies a 2D calculation for one uniform axial region with reflecting boundaries at the top and bottom of the pin.

Three potential axial region distributions were studied. The first was a simple linear distribution (evenly distributing the axial regions along the fuel rod axis). This distribution is given by:

$$z(j)=\frac{H}{2}\frac{j}{N_z} \qquad for \quad j=0,1,2,\ldots,N_z \tag{6}$$

where $z(j)$ is the upper boundary of axial fuel region j, H is the total rod height, and N_z is the total number of axial fuel regions for each pin.

The second distribution considered was selected to allow the axial fuel regions to assume a cosine shape. This distribution is given by:

$$z(j)=\frac{H}{2}\left[1-\cos\left(\frac{\pi}{2}\frac{j}{N_z}\right)\right] \qquad for \quad j=0,1,2,\ldots,N_z. \tag{7}$$

This distribution is reminiscent of the power profile in the pin for a homogeneous reactor.

The last distribution is similar to that used for the radial fuel regions in Eq. (5). This distribution is an exponential distribution in which many more regions are placed near the ends of the pin than near the center. The exponential distribution can be calculated using the following:

$$z(j)=\frac{H}{2}\left[1-\frac{1-\exp(-\Sigma_a j)}{1-\exp(-\Sigma_a N_r)}\right] \qquad for \quad j=1,2,3,\ldots,N_r \qquad (8)$$

where Σ_a is a one-group macroscopic absorption cross section and the other variables are the same as those mentioned above. As mentioned before, for PWRs, the one-group cross section can be well approximated using the ^{235}U and ^{238}U thermal absorption cross section values. Thus, for 3.0 w/o ^{235}U fuel, $\Sigma_a \approx 0.55$ cm^{-1}. This value will change depending on the spectrum of neutrons in the reactor which is affected by the temperature, the fuel composition, and the fuel density; however, for most LWR reactors, using a one-group cross section of 0.55 cm^{-1} yields a reasonable axial fuel region distribution.

To determine which axial distribution yielded the best results with the fewest number of regions, calculational experiments were performed with Monteburns using 0, 1, 2, 3, 4, 5, 6, and 12 fuel regions for each type of axial region distribution. It was determined that there was less than a 0.02% difference between the 6 region case using the exponential distribution and the 12 region case using the exponential distribution. It was therefore assumed that the 12 region case using the exponential distribution

closely approximated the fine mesh limit. Note that in order to acquire a sufficient number of neutron interactions near the ends of the pins, the total number of histories used in the axial distribution calculations was increased to 6.4 MH. This number of histories appears to be the minimum required to achieve sufficiently small statistical uncertainties at higher burnups in the end regions of the pin.

Comparisons were made for each axial region distribution for all of the desired isotopic ratios. It was determined that using an exponential distribution of the axial fuel regions yielded a result indistinguishable from the fine mesh limit, with the fewest number of fuel regions compared to the other proposed distributions. In all cases, the MPDs for the exponential distribution decreased more rapidly than for the other distributions. The MPD results for the axially-averaged isotopic ratios for the exponential axial region distributions are listed in Table IV.

As can be seen from the values in Table IV, all of the non-actinide MPDs are relatively insensitive to the axial profile employed. Thus, when performing spent fuel studies in PWRs, it is only necessary to have a detailed axial profile for determining actinide concentrations. For studies interested only in axially-averaged fission product concentrations, such as the environmental monitoring study proposed here, a 2D calculation is generally sufficient. Since actinides are not a major consideration here, a 2D pin cell with nine flat-flux radial fuel regions and a 2000 MWd/MTU burnup step size was used to determine the results in the remainder of this section.

TABLE IV

Maximum Percent Deviation of Various Isotopic Ratios from the Value at the Fine Mesh Limit (Chosen as 14 Axial Regions) for Different Numbers of Axial Regions

	Maximum Percent Deviation from Fine Mesh Limit (%)						
# of Fuel Regions	$^{235}U/^{238}U$	$^{240}Pu/^{239}Pu$	$^{83}Kr/^{86}Kr$	$^{84}Kr/^{86}Kr$	$^{131}Xe/^{134}Xe$	$^{132}Xe/^{134}Xe$	$^{136}Xe/^{134}Xe$
0*	1.97	1.26	0.77	0.25	0.50	0.10	0.15
1	1.87	1.24	0.54	0.21	0.45	0.08	0.12
2	1.06	0.75	0.15	0.06	0.24	0.04	0.06
3	0.52	0.15	0.03	**	0.07	**	**
4	0.22	0.15	**	**	**	**	**
5	0.09	0.06	**	**	**	**	**
6	**	**	**	**	**	**	**

* A value of zero implies the calculation is performed for an axially uniform (or 2D) pin cell.

** Accurate to within five significant digits.

5. Power Density: Neutron Flux Level

One parameter that varies considerably for reactors (power, production, or research reactors) is the power level. Changes in power would occur during startup, shutdown, reloading, changing of targets, or changing of experimental devices. The power could also be altered to adjust for load conditions or to decrease (or increase) certain isotope production rates (for instance, in a research reactor). The spent fuel isotopic concentrations are affected by the power level through two processes. The first process is through a change in temperature which consequently alters the neutron spectrum and interaction cross sections used in the depletion calculations. The second process is through a change in the time at power to reach a specific burnup. This leads

to longer (or shorter) decay times for parent isotopes, and consequently can alter the buildup and depletion of fission products and actinides (specifically the plutonium isotopes).

The effect of neutron flux level on the desired isotopic ratios was investigated by simulating the model PWR at various power levels. For the purposes of the investigations here, it was assumed that the average fuel temperature and coolant temperature remained constant and only the neutron flux level was changed. Effects from changes in material temperatures will be studied in a later section. Calculations were performed at a specific power of 15.0, 20.0, 25.0, 30.0, 35.0, 40.0, and 45.0 W/g using a 2D modified pin cell with nine flat-flux fuel regions and a 2000 MWd/MTU burnup step size. Comparisons between the isotopic ratios of interest for the various power levels were made using the 30.0 W/g results as the nominal value. The MPDs from the 30.0 W/g values are listed in Table V.

For most isotopes the power level has a negligible effect; however, for the $^{235}U/^{238}U$, $^{240}Pu/^{239}Pu$, and $^{136}Xe/^{134}Xe$ isotopic ratios the effect is significant. It is postulated that the actinide effects are primarily due to the decay of parent isotopes (specifically ^{239}Np). The ^{136}Xe concentration is significantly affected by the production from ^{135}Xe. The absorptions in ^{135}Xe vary depending upon the neutron flux level and the decay time of its parent isotope ^{135}I. Thus, for investigations concerning actinide isotopes or fission products with complicated decay and absorption chains, changes in neutron flux level (and consequently power density) may prove important. Previous studies have come to similar conclusions.[49]

TABLE V
Maximum Percent Deviation of Various Isotopic Ratios from the Value at Nominal Power Density (Chosen as 30.0 W/g) for Several Specific Power Levels

	Maximum Percent Deviation from Fine Mesh Limit (%)						
Power Level (W/g)	$^{235}U/^{238}U$	$^{240}Pu/^{239}Pu$	$^{83}Kr/^{86}Kr$	$^{84}Kr/^{86}Kr$	$^{131}Xe/^{134}Xe$	$^{132}Xe/^{134}Xe$	$^{136}Xe/^{134}Xe$
15.0	1.48	10.47	0.24	0.13	0.04	0.39	8.81
20.0	0.74	6.85	0.16	0.07	0.04	0.27	4.90
25.0	0.28	3.38	0.09	0.04	0.03	0.14	2.13
30.0	0.00	0.00	0.00	0.00	0.00	0.00	0.00
35.0	0.21	3.28	0.08	0.03	0.02	0.14	1.65
40.0	0.32	6.41	0.09	0.04	0.03	0.26	2.93
45.0	0.37	9.34	0.12	0.08	0.05	0.39	3.96

6. Power Density: Fuel and Moderator Temperature

To examine the affect of different fuel and moderator temperatures on the aforementioned isotopic ratios, cross section sets at five fuel temperatures (400K, 700K, 800K, 900K, and 1200K) and five moderator temperatures (350K, 500K, 550K, 600K, and 750K) were produced using NJOY.[50] The cross section sets included the most important actinides (^{235}U, ^{236}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu), the fission products of interest (^{131}Xe, ^{132}Xe, ^{134}Xe, ^{136}Xe, ^{83}Kr, ^{84}Kr, ^{85}Kr, and ^{86}Kr), and the moderator isotopes (^{1}H and ^{16}O). All other isotopes were evaluated at 300K (the default MCNP cross section sets). The PWR from Table I was then simulated using a 2D modified pin cell with nine flat-flux fuel regions and a 2000 MWd/MTU burnup step size for burnups from 0 to 50000 MWd/MTU. Comparisons between the isotopic

ratios of interest for the various fuel temperatures were made using the 800K results as the nominal value. The MPDs from the 800K values are listed in Table VI.

Comparisons between the isotopic ratios of interest for the various moderator temperatures were made using the 550K results as the nominal value. The MPDs from the 550K values are listed in Table VII. The MPDs resulting from changes in the fuel temperature are generally small for non-actinide isotopes even when the temperature of the fuel is changed by a factor of two. A similar result is found for changes in the moderator temperature. For this reason, small changes in the fuel temperature resulting from a slowly varying power distribution (such as is found in a large reactor) will have negligible effects on the isotopic ratios of interest. Thus, small changes in moderator and fuel temperature will be neglected in the models developed below.

TABLE VI
Maximum Percent Deviation of Various Isotopic Ratios for Several Fuel Temperatures from the Value Calculated at 800K

	Maximum Percent Deviation from Fine Mesh Limit (%)						
Temperature (K)	$^{235}U/^{238}U$	$^{240}Pu/^{239}Pu$	$^{83}Kr/^{86}Kr$	$^{84}Kr/^{86}Kr$	$^{131}Xe/^{134}Xe$	$^{132}Xe/^{134}Xe$	$^{136}Xe/^{134}Xe$
400	12.95	3.70	2.69	0.59	0.98	0.21	0.44
700	3.01	1.08	0.62	0.14	0.24	0.05	0.13
800	0.00	0.00	0.00	0.00	0.00	0.00	0.00
900	2.90	1.02	0.54	0.13	0.24	0.05	0.13
1200	11.15	3.70	2.70	0.48	0.83	0.17	0.38

TABLE VII
Maximum Percent Deviation of Various Isotopic Ratios for Several Moderator Temperatures from the Value Calculated at 550K

	Maximum Percent Deviation from Fine Mesh Limit (%)						
Temperature (K)	$^{235}U/^{238}U$	$^{240}Pu/^{239}Pu$	$^{83}Kr/^{86}Kr$	$^{84}Kr/^{86}Kr$	$^{131}Xe/^{134}Xe$	$^{132}Xe/^{134}Xe$	$^{136}Xe/^{134}Xe$
350	12.13	19.23	1.60	1.18	0.48	0.38	0.57
500	3.22	5.71	0.46	0.34	0.15	0.11	0.26
550	0.00	0.00	0.00	0.00	0.00	0.00	0.00
600	3.12	5.71	0.45	0.32	0.14	0.10	0.26
750	11.74	23.07	1.59	1.17	0.51	0.38	1.15

7. Reactivity Control

Several parameters can be manipulated to control the reactivity in a reactor. These include, but are not limited to, the addition of burnable absorbers, the movement of control rods, and the increase/decrease in chemical shim concentration. Burnable absorber rods are added to an assembly to decrease the assembly reactivity at low burnups. The effect of burnable absorbers and control rods is left for later investigations.

Studies were performed to determine the effect of chemical shim (soluble boron concentrations) in the coolant water on spent fuel isotopic compositions. The desired isotopic ratios were calculated for the model PWR with constant values of 0, 200, 400, 600, 800, 1200, 1600, and 3200 ppm natural boron in the water for burnups from 0 to 50000 MWd/MTU. The MPDs for the 200, 400, 600, 800, 1200, 1600, and 3200 ppm cases relative to the 0 ppm case are listed in Table VIII. As can be seen, the effect on

the actinides is considerable even at low boron concentrations. This suggests that calculations should be performed with reactivity control (or using the critical spectrum).

To be rigorously correct, any steady-state calculation should make use of the critical spectrum. Monteburns does not perform calculations using the critical spectrum, thus investigations were performed using the HELIOS lattice-physics code[17] to determine the effect of the use of the critical spectrum on the isotopic inventories of spent fuel. To determine the effect of using the critical spectrum, calculations were performed for the model PWR using the HELIOS lattice-physics code with and without the critical spectrum. The percent deviations of the selected isotopic ratios calculated without the critical spectrum from those calculated with the critical spectrum are listed in Table IX.

TABLE VIII
Maximum Percent Deviation of Various Isotopic Ratios from the Value Calculated With No Soluble Boron for Various Boron Concentrations (in ppm)

	Maximum Percent Deviation from Fine Mesh Limit (%)						
Boron Conc. (ppm)	$^{235}U/^{238}U$	$^{240}Pu/^{239}Pu$	$^{83}Kr/^{86}Kr$	$^{84}Kr/^{86}Kr$	$^{131}Xe/^{134}Xe$	$^{132}Xe/^{134}Xe$	$^{136}Xe/^{134}Xe$
200	2.99	1.32	0.61	0.15	0.07	0.11	0.19
400	5.98	2.59	1.15	0.29	0.10	0.21	0.32
600	8.92	3.81	1.69	0.42	0.16	0.30	0.45
800	11.81	4.98	2.15	0.56	0.19	0.41	0.57
1200	17.57	7.22	3.15	0.84	0.29	0.59	0.89
1600	23.18	9.31	4.07	1.09	0.38	0.79	1.15
3200	44.50	16.40	7.06	2.03	0.73	1.48	2.23

TABLE IX
Percent Deviation of Various Isotopic Ratios Calculated Without Using the Critical Spectrum from the Values Calculated Using the Critical Spectrum (Calculations Performed Using the HELIOS Lattice-Physics Code[17])

Isotopic Ratio	Percent Deviation (%)
$^{235}U/^{238}U$	1.48
$^{240}Pu/^{239}Pu$	1.81
$^{83}Kr/^{86}Kr$	0.23
$^{84}Kr/^{86}Kr$	0.37
$^{131}Xe/^{134}Xe$	0.19
$^{132}Xe/^{134}Xe$	0.19
$^{136}Xe/^{134}Xe$	0.14

The results in Table IX suggest that use of the critical spectrum (and consequently correct reactivity control) in calculating spent fuel isotopics may be crucial for actinide isotopes, but the effect is less significant for the fission product concentrations. It should be noted that for the model PWR with burnups from 0 to 50000 MWd/MTU, the infinite multiplication factor for the assembly (as calculated by HELIOS) ranged from 1.3599 to 0.7770. Because Monteburns is incapable of performing critical spectrum calculations (without major modifications), this effect will be neglected in future calculations.

8. Suggested Model

The sensitivity of certain isotopic ratios to various reactor parameters has been studied and reported above. These studies suggested the use of a 2D modified pin cell

with nine flat-flux radial fuel regions and a 2000 MWd/MTU burnup step size for accurately determining the fission product isotopic ratios of interest. The pin-to-pin pitch of the modified pin cell can be calculated using Eq. (1). The radial fuel region distributions can be calculated using Eq. (5). It was determined that small changes in the power density and critical spectrum effects could be neglected for all of the non-actinide isotopic ratios. This model is expected to be adequate for use in the environmental monitoring application.

B. Code Selection

The next step in these calculations was to choose a code system that was well suited to calculating noble gas isotopic ratios in spent fuel from a variety of reactors. Access to a set of reactor analysis codes was established. These codes included the HELIOS lattice-physics code[17], a linked MCNP-4B/ORIGEN 2.1 code named Monteburns 3.01[39], the comprehensive code package WIMS-7A[51], the point-depletion code ORIGEN 2.1[41], a linked DANT[52]/CINDER[53] code system, and the Oak Ridge National Laboratory analysis package SCALE 4.3[54]. The most suitable code for the environmental monitoring application was chosen based on the following four characteristics:

1. Capability (the ability to calculate concentrations for all isotopes of interest),
2. Accuracy (the ability to accurately predict noble gas isotopic ratios),
3. Versatility (the ability to handle numerous kinds of reactors),

4. Computational speed (the ability to perform the calculations quickly).

A series of experimental data taken from the literature was used to judge each code system's adherence to these criteria. These benchmarks also allowed for the determination of the degree of accuracy of the calculations and the resultant database.

1. Benchmark Data

A literature search was performed to uncover all available experimental data pertaining to measurements of fission product and heavy metal isotopic concentrations in spent fuel. This search resulted in a set of twelve different reactors for which destructive spent fuel examinations had been reported. The twelve reactors, along with the reactor type and the information reported for the experiments, are listed in Table X.

As can be seen, there exist numerous experiments on light-water moderated, power reactors (LWRs); however, few experiments were found containing results for heavy-water moderated, graphite-moderated, or fast reactors. Also, there was no data available in the open-literature for production reactor fuel. For this reason, the comparisons performed here will be primarily related to LWRs, and extrapolation to other reactor types may require further validation of the code system.

The Borssele, Calvert Cliffs, Genkai, Mihama, Obrigheim, and Turkey Point reactors represent a set of fairly standard power PWRs. Each uses square, uranium dioxide fueled assemblies with B_4C control rods, minimal burnable absorbers, and single enrichment fuel pins. On the other-hand, the Shippingport and Trino Vercellese reactors are not of the standard PWR variety. Shippingport is a fully enriched (93%)

uranium driver and natural uranium blanket, light water moderated and cooled, pressurized water reactor with plate fuel. The Trino Vercellese reactor's core geometrically resembles a BWR (i.e., fuel assemblies containing multiple enrichment pins and cruciform type control rods) even though the coolant is maintained under pressure during operation and not allowed to boil. The Trino Vercellese fuel assemblies are oddly shaped consisting of 208 rods per assembly with spaces for the cruciform control rods. The control rods are fuel followed with 2.73 w/o ^{235}U fuel.

TABLE X
Reactor Systems from the Literature Used in Reactor Physics Code Benchmarking

Reactor Name	Reactor Type	Burnup Range (MWd/MTU)	Heavy Metal Isotopics Reported	Noble Gas Isotopics Reported
Borsselle[55]	PWR	31,000–32,000		✔
Calvert Cliffs[56-58]	PWR	18,680–46,460	✔	✔
Garigliano[59]	BWR	9,800–14,480	✔	✔
Genkai[44]	PWR	29,440–38,700	✔	✔
Gundremmingen[60]	BWR	14,390–25,900	✔	✔
Halden OECD[61-63]	HBWR	15,040–28,600		✔
Mihama[44]	PWR	6,900–21,200	✔	✔
Obrigheim[64]	PWR	15,600–36,260	✔	✔
Pickering[65]	CANDU	9,200	✔	
Shippingport[66]	PWR	2,414–22,000	✔	
Trino Vercellese[67,68]	PWR	3,399–27,758	✔	✔
Turkey Point[69]	PWR	30,510–31,560	✔	✔

Gundremmingen and Garigliano are fairly standard BWRs, both employing low enriched uranium dioxide fuel. Gundremmingen is composed of 6×6 BWR fuel assemblies; however, Garigliano uses GE 9×9 assemblies. Both have cruciform type control rods and coolant channels. The only non-standard aspect of these cores was their rather low enrichment (1.6-2.5 w/o ^{235}U).

The Pickering Generating Station uses CANDU-600's. These 600 MWe reactors are heavy water moderated and cooled. They are fueled with natural uranium dioxide fuel in Zircaloy cladding. The CANDU-600 is representative of most of the world's CANDU reactors.

The Halden OECD Reactor is a heavy water moderated, boiling water reactor. It is primarily used for research and experimental purposes. The reactor is composed of seven element fuel bundles in a triangular lattice with 1.5 w/o ^{235}U uranium dioxide fuel. The reactor is cooled by heavy water flowing in cooling channels around the fuel bundles; however, the primary source of moderator is a large tank of subcooled heavy water.

For detailed information regarding the geometry and operating characteristics of the benchmark experiment reactors see the *Directory of Nuclear Reactors*,[70] the *Directory of Nuclear Power Plants Around the World*,[71] and the individual experimental reports referenced in Table X.

All of the measurements performed for spent fuel from the reactors listed in Table X were mass spectrometric measurements on fuel samples cut from fuel pins from assemblies (except for the Borselle results which were for complete assembly

dissolutions). The fuel samples were from various heights in the reactor cores and were not necessarily representative of the assembly average. Also, the uncertainties for these measurements ranged from ±1% to ±6%; however, the vast majority of the measurements had reported uncertainties of around ±1-2%.

2. Benchmarking Results

Each of the reactors in Table X were modeled using the following reactor analysis codes: HELIOS, Monteburns 3.01, SCALE 4.3, DANT/CINDER, and ORIGEN 2.1. For each of these reactors, a model based on the methodology described in Section III.A.8. above was used (i.e., a 2D modified pin cell with nine flat-flux radial fuel regions and a 2000 MWd/MTU burnup step-size), for all of the codes (except the point code ORIGEN 2.1) to determine the xenon and krypton isotopic concentration ratios in the fuel at each burnup step. These values were then compared to the reported values from the literature.

Figures 15-18 contain the xenon and krypton ratio results found for the Obrigheim reactor with 3.00 w/o ^{235}U fuel. As can be seen, the Monteburns code system results in predicted isotopic ratios versus burnup which more closely matches that of the reported measurements than any other code system for all of the isotopic ratios of interest. Similar results are found for the other reactors. Isotopic ratio plots (similar to Figs. 15-18) for the other reactors from the literature are included in Appendix A.

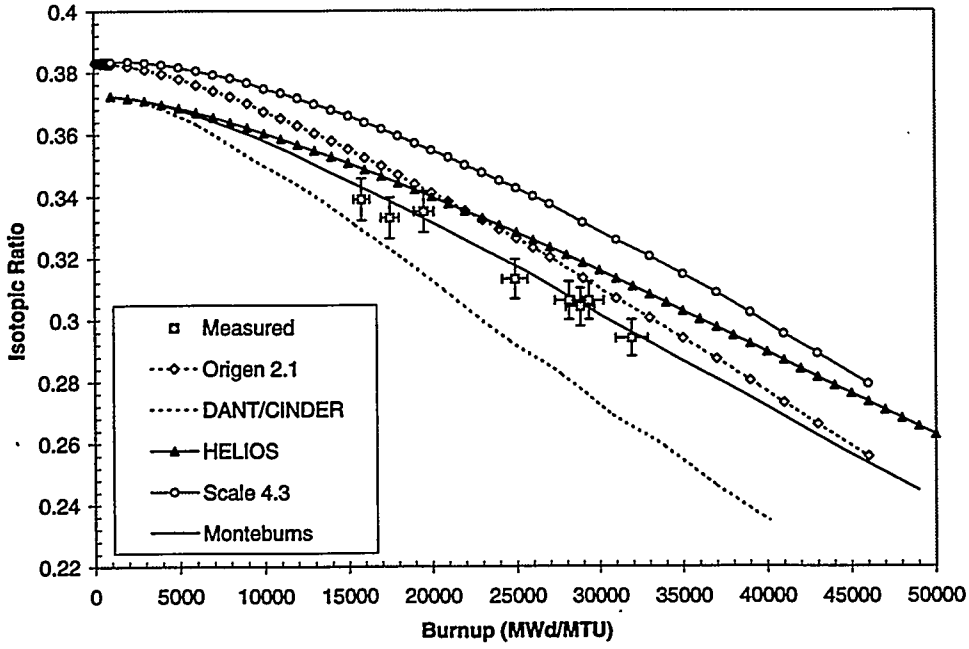

Fig. 15. $^{131}Xe/^{134}Xe$ isotopic ratio versus burnup for Obrigheim 3.00 w/o ^{235}U fuel.

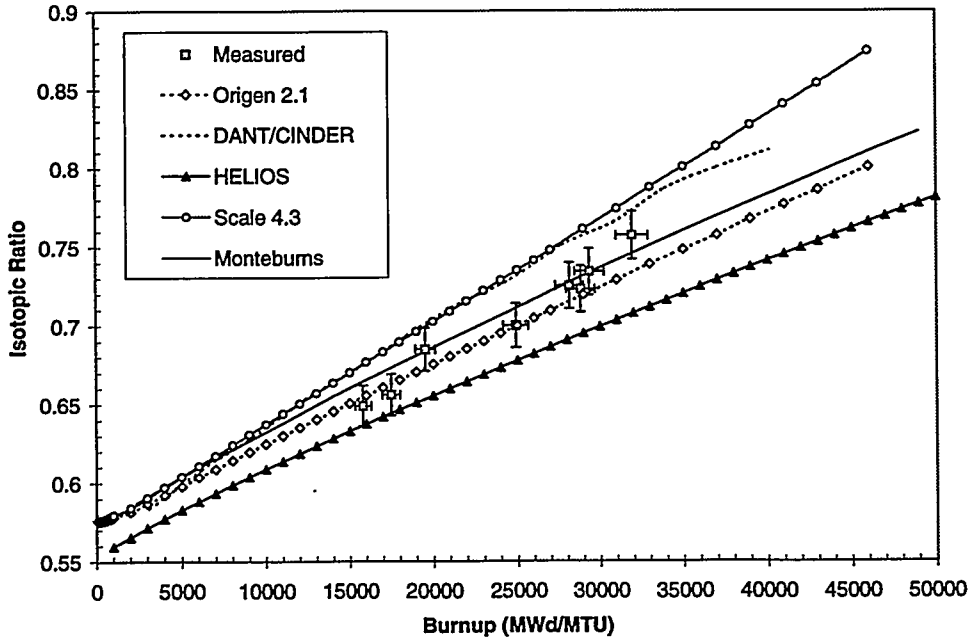

Fig. 16. $^{132}Xe/^{134}Xe$ isotopic ratio versus burnup for Obrigheim 3.00 w/o ^{235}U fuel.

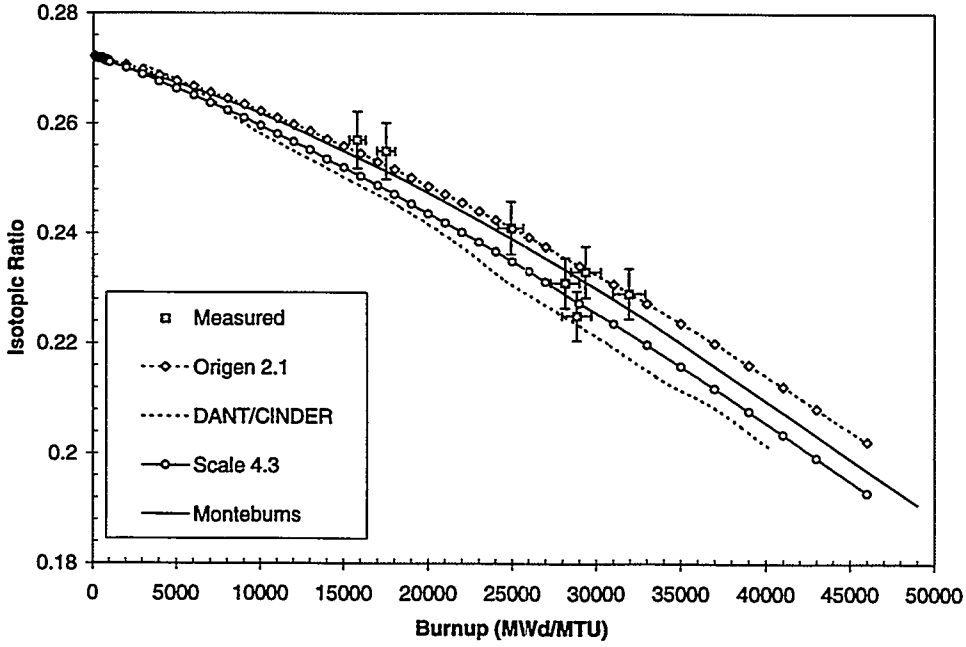

Fig. 17. $^{83}Kr/^{86}Kr$ isotopic ratio versus burnup for Obrigheim 3.00 w/o ^{235}U fuel.

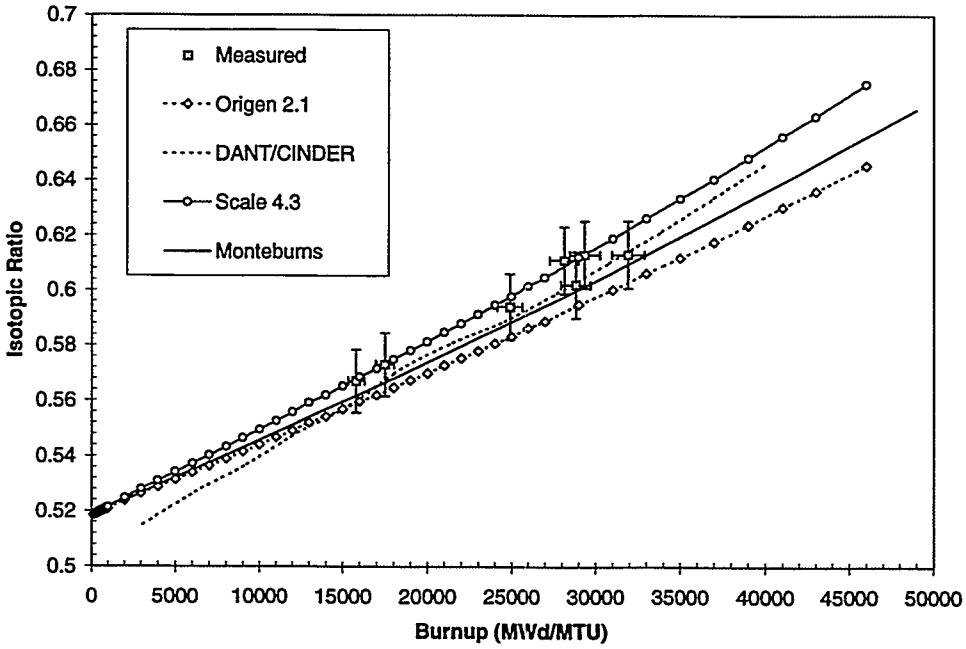

Fig. 18. $^{84}Kr/^{86}Kr$ isotopic ratio versus burnup for Obrigheim 3.00 w/o ^{235}U fuel

TABLE XI
Benchmark Statistics for Each Code System

	Average Percent Difference Between Measured and Calculated Isotopic Ratios			
Analysis Code	$^{131}Xe/^{134}Xe$	$^{132}Xe/^{134}Xe$	$^{83}Kr/^{86}Kr$	$^{84}Kr/^{86}Kr$
Monteburns	0.72	0.97	1.02	0.66
ORIGEN 2.1	3.13	1.31	1.09	2.18
HELIOS	3.34	3.25	n/a	n/a
DANT/CINDER	5.31	3.28	2.89	0.89
SCALE 4.3	6.81	3.50	2.11	0.66

Note that the WIMS-7A code was not used in these analyses because the WIMS data libraries did not include cross section and fission yield data for ^{84}Kr, ^{85}Kr, ^{86}Kr, ^{134}Xe, and ^{136}Xe. Also, the HELIOS code was used only for the xenon isotopes, since its data libraries did not include fission yields for the krypton isotopes. ORIGEN 2.1 is a point depletion code so the model used was very simple and did not require pin cell geometric specifications. Also, ORIGEN 2.1 was not capable of modeling the Halden OECD reactor since there were no heavy water moderated, boiling water reactor (HBWR) data libraries available.

The benchmark data produced was compiled by analyzing the average percent difference between the measured and calculated isotopic ratios for each code and for each ratio of interest (specifically $^{83}Kr/^{86}Kr$, $^{84}Kr/^{86}Kr$, $^{131}Xe/^{134}Xe$, and $^{132}Xe/^{134}Xe$). The average percent differences are listed in Table XI. The Monteburns code system is

more accurate than all of the other code systems at predicting the isotopic ratios of interest for all of the reactors analyzed. Table XI also suggests that ORIGEN 2.1 is fairly accurate at predicting these isotopic ratios; however, this may be misleading since ORIGEN 2.1 is incapable of modeling reactors for which it does not have a data library (such as for RBMK reactors).

3. Code Selection and Estimation of Accuracy

A reactor analysis code was selected for use with the environmental monitoring system based on the selection criteria specified above. Each code considered was ranked according to its capability to solve the problem at hand, its accuracy at predicting the isotopic ratios of interest, its versatility in analyzing many reactor types, and its computational speed. The results from this ranking are displayed in Table XII. From this table, it is found that the Monteburns code system is superior to the other codes analyzed for capability, accuracy, and versatility; however, the code is extremely computationally intensive. The HELIOS code is also a reasonable choice except that it is unable to calculate the krypton isotopic ratios without significant modification to the existing libraries. Also, its flux solution tends to diverge when analyzing fast reactor systems.

TABLE XII
Adherence to Selection Criteria for Each Code System Analyzed

Code System	Capability	Accuracy	Versatility	Speed
Monteburns	Excellent	Excellent	Excellent	Poor
ORIGEN 2.1	Excellent	Good	Poor	Excellent
DANT/CINDER	Excellent	OK	Excellent	Good
HELIOS	OK	OK	Good	Excellent
SCALE 4.3	Excellent	Poor	Poor	Good
WIMS-7A	Poor	Undetermined	OK	Good

The DANT/CINDER code system might be a reasonable choice, but to date it does not exist in a fully integrated package (i.e., cross section processing, flux solution, and burnup module). Also, the accuracy of the DANT/CINDER code appears to be significantly less than that of Monteburns. However, with some development, it may be used more effectively in the future.

ORIGEN 2.1 is fast computationally and is capable of calculating all fission products and actinides of interest; however, the code uses prebuilt neutron flux spectra, fission yields, and cross section libraries for a limited set of reactors. For this reason, its versatility was labeled as poor. Also, there exists some question as to whether the code is accurate in the low burnup regions. Since ORIGEN 2.1 uses prebuilt libraries that have been modified to yield accurate answers at a specific burnup point (33000 MWd/MTU for PWRs), the code does not have extensive physics built in. The modification of these libraries may lead to a significant error when far from the

libraries design point (i.e., very low or very high burnup). The environmental monitoring system developed here must be accurate for an extensive range of burnups; thus, it may be questionable whether ORIGEN 2.1 alone can provide the accuracy needed.

The two other code packages considered (WIMS-7A and SCALE 4.3) have serious difficulties when confronting this problem. The WIMS-7A data libraries do not include most of the fission products of interest, and adding these isotopes to the libraries is beyond the scope of this project. SCALE 4.3 is unable to model any but the simplest of reactor types (i.e., PWRs and BWRs) and its accuracy even for these types may not be sufficient for the current application.

This reasoning led to the conclusion that the Monteburns code system was the appropriate choice for generating the reactor physics database (primarily due to its superior nuclear data). The poor computational speed of Monteburns was overcome by using its flux solver (MCNP-4B) in PVM (parallel virtual machine) mode. Since MCNP is a Monte Carlo code, it is well suited for calculations in a parallelized environment. This allows for the linking of numerous processors to carry out separate tasks (i.e., track particle histories) and greatly increases the computational speed of Monteburns. In general, it was determined that a single pin cell in Monteburns using 10 processors required approximately 36 hours of computational time (using 3.2 MH). However, the code system resulted in xenon and krypton isotopic ratios consistently accurate to within ±1%, and the code had the ability to model any reactor system in existence today.

C. Database Creation

Reactor physics databases for the Inverse and Forward Problems discussed in Section II were generated using the Monteburns 3.01 code system. For the Inverse Problem, a database of xenon and krypton isotopic ratios as a function of burnup for fourteen different reactor fuel types was constructed. The reactor fuel types in this database (listed in Table XIII) formed an exhaustive, mutually exclusive set of reactor types. Each fuel type in this database was developed such that it would provide a representative average of all of the reactors of that type. This averaging is described in more detail below. The database for the Forward Problem consisted of as many different fuel types as time allowed. These included all of the fuels listed in Table XIV; however, more fuels may be added in the future as the need for them becomes apparent.

Each of the databases was created in Microsoft Access and contained, in addition to the xenon and krypton isotopic ratios, plutonium concentrations (in g/MTHM), $^{240}Pu/^{239}Pu$ isotopic ratios, and various fission product concentrations (in g/MTHM) versus burnup. The exact information stored in the databases is shown in Table XV. This information encompasses an extensive range of burnups, and even though the methodology described above suggested that the usage of a 2000 MWd/MTU burnup step-size was sufficient, the models were executed using a 1000 MWd/MTU burnup step-size to allow for more accurate interpolations.

TABLE XIII
Reactor Models Included in the Inverse Problem Reactor Physics Database

Reactor Model	Description
PWR-LEU	Pressurized water reactor with 2.85 w/o ^{235}U uranium dioxide fuel in a Zircaloy clad.
PWR-MOX	Pressurized water reactor containing 30% 35,000 MWd/MTU Pu recycle and the remainder 3.00 w/o ^{235}U uranium dioxide fuel.
BWR-LEU	Boiling water reactor with 2.75 w/o ^{235}U uranium dioxide fuel in a Zircaloy clad.
BWR-MOX	Boiling water reactor containing 30% 35,000 MWd/MTU Pu recycle and the remainder 2.85 w/o ^{235}U uranium dioxide fuel.
CANDU-Nat.U	Heavy water moderated, heavy water cooled, natural uranium fuel reactor.
CANDU-Dupic	Heavy water moderated, heavy water cooled reactor with 35,000 MWd/MTU Pu recycle.
LMFBR Driver	Sodium cooled, fast reactor with 70% enriched uranium metal fuel.
LMFBR Blanket	Sodium cooled, fast reactor with depleted uranium metal targets.
RBMK	Graphite moderated, water cooled reactor with slightly enriched uranium dioxide fuel.
Calder Hall	Graphite moderated, carbon dioxide cooled reactor with natural uranium fuel.
Hanford-N	Graphite moderated, water cooled reactor with natural uranium metal fuel.
HTGR	Graphite moderated, helium cooled reactor with thorium and highly enriched uranium fuel.
U.S. Production Reactor Target	Heavy water moderated, heavy water cooled with depleted uranium targets.
U.S. Production Reactor Driver	Heavy water moderated, heavy water cooled with enriched uranium driver fuel.

TABLE XIV
Reactor Models Included in the Forward Problem Reactor Physics Database

Reactor Model	Description
Westinghouse 15×15 (3.00 w/o)	PWR with 3.00 w/o ^{235}U in Zircaloy clad.
Westinghouse 17×17 (3.00 w/o)	PWR with 3.00 w/o ^{235}U in Zircaloy clad.
Westinghouse 17×17 (3.20 w/o)	PWR with 3.20 w/o ^{235}U in Zircaloy clad.
Siemens 14×14 (2.85 w/o)	PWR with 2.85 w/o ^{235}U in Zircaloy clad.
Mitsubishi 15×15 (3.00 w/o)	PWR with 3.00 w/o ^{235}U in Zircaloy clad.
GE 6×6 (2.50 w/o)	BWR with 2.50 w/o ^{235}U in Zircaloy clad.
GE 8×8 (2.75 w/o)	BWR with 2.75 w/o ^{235}U in Zircaloy clad.
RBMK 1500	RBMK with 2.4 w/o ^{235}U fuel.
Calder Hall	Calder Hall with natural uranium fuel.
Ft. Saint Vrain	HTGR with thorium and highly enriched fuel
EBR-II Target Fuel	LMFBR depleted uranium target fuel.
EBR-II Driver Fuel	LMFBR highly enriched (70%) driver fuel.
U.S. Production Reactor Target	Depleted uranium target fuel.
U.S. Production Reactor Driver	Highly enriched driver fuel.
CANDU 600	600 MWe CANDU (natural uranium fueled).
CANDU 800	800 MWe CANDU (natural uranium fueled).

TABLE XV
Isotopic Information Contained in the Databases for Each Reactor Model and at Each Burnup Point

Information	Units	Usage
$^{130}Xe/^{134}Xe$	isotopic ratio	burnup and fuel type determination
$^{131}Xe/^{134}Xe$	isotopic ratio	burnup and fuel type determination
$^{132}Xe/^{134}Xe$	isotopic ratio	burnup and fuel type determination
$^{136}Xe/^{134}Xe$	isotopic ratio	operational history determination
$^{82}Kr/^{86}Kr$	isotopic ratio	burnup and fuel type determination
$^{83}Kr/^{86}Kr$	isotopic ratio	burnup and fuel type determination
$^{84}Kr/^{86}Kr$	isotopic ratio	burnup and fuel type determination
$^{85}Kr/^{86}Kr$	isotopic ratio	fuel age determination
^{238}Pu	g/MTHM	extra information
^{239}Pu	g/MTHM	extra information
^{240}Pu	g/MTHM	extra information
^{241}Pu	g/MTHM	extra information
^{242}Pu	g/MTHM	extra information
$^{238}Pu/^{239}Pu$	isotopic ratio	proliferation indicator
^{237}Np	g/MTHM	waste hazard
^{241}Am	g/MTHM	waste hazard
^{242m}Am	g/MTHM	waste hazard
^{243}Am	g/MTHM	waste hazard
^{242}Cm	g/MTHM	waste hazard
^{243}Cm	g/MTHM	waste hazard
^{244}Cm	g/MTHM	waste hazard
^{245}Cm	g/MTHM	waste hazard
^{134}Cs	g/MTHM	waste hazard
^{135}Cs	g/MTHM	burnup monitor
^{137}Cs	g/MTHM	burnup monitor
^{99}Tc	g/MTHM	waste hazard
^{129}I	g/MTHM	waste hazard
^{90}Sr	g/MTHM	waste hazard
^{146}Nd	g/MTHM	burnup monitor
^{148}Nd	g/MTHM	burnup monitor

Results for each of the models listed in Table XIII were generated using an appropriate average model for that reactor type (except for the production reactor target and driver fuels, which were modeled explicitly). This average model was determined from an extensive representative set of reactors for that type. For example, for the PWRs, a set of twelve different reactor fuels was modeled and the representative average was generated by numerical experimentation until an appropriate average was constructed. The $^{131}Xe/^{134}Xe$ and $^{132}Xe/^{134}Xe$ isotopic ratios for the set of PWR fuel used and the representative average is shown in Figs. 19 and 20. As can be seen, the average PWR model represents a good mean for the set. A similar procedure was used for the generation of the representative average models for the other reactor types. Some of the Monteburns 3.01 input decks used for the construction of these databases are included in Appendix B.

The construction of the model averages also aided in the estimation of the standard deviation of the calculated isotopic ratios using the spread of these reactors about the representative average. These standard deviations were generated for each burnup point in the database using the well known equation for estimating the standard deviation:

$$\sigma^2 = \frac{\sum_{i=1}^{N}(x_i - \mu)^2}{N-1} \qquad (9)$$

where σ is the standard deviation, x_i is the value of the isotopic ratio at the given burnup point for model i, μ is the value of the (sample) average isotopic ratio at the

given burnup point, and N is the total number of models used. For the representative PWR, the fractional standard deviations of the isotopic ratios were generally ~1.5%.

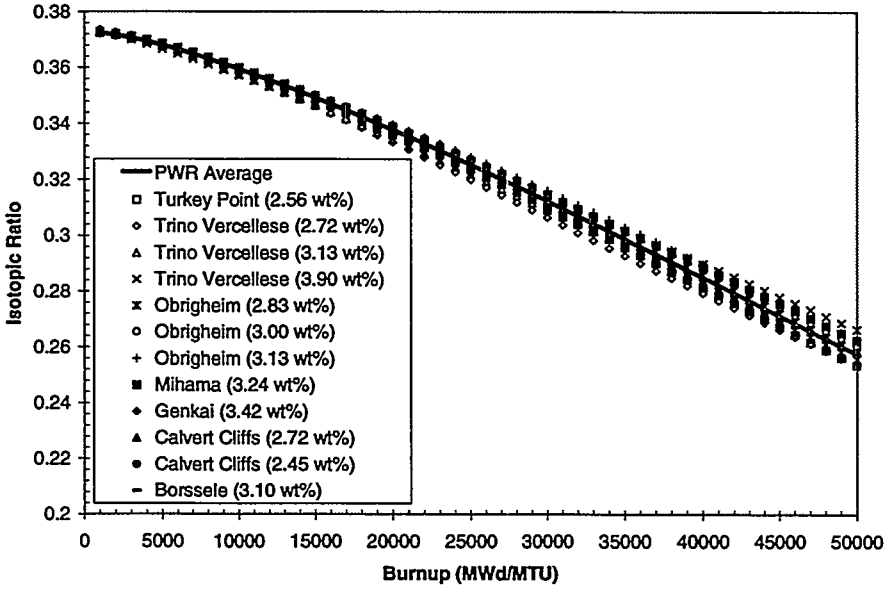

Fig. 19. $^{131}Xe/^{134}Xe$ isotopic ratio versus burnup for several PWRs and for average PWR.

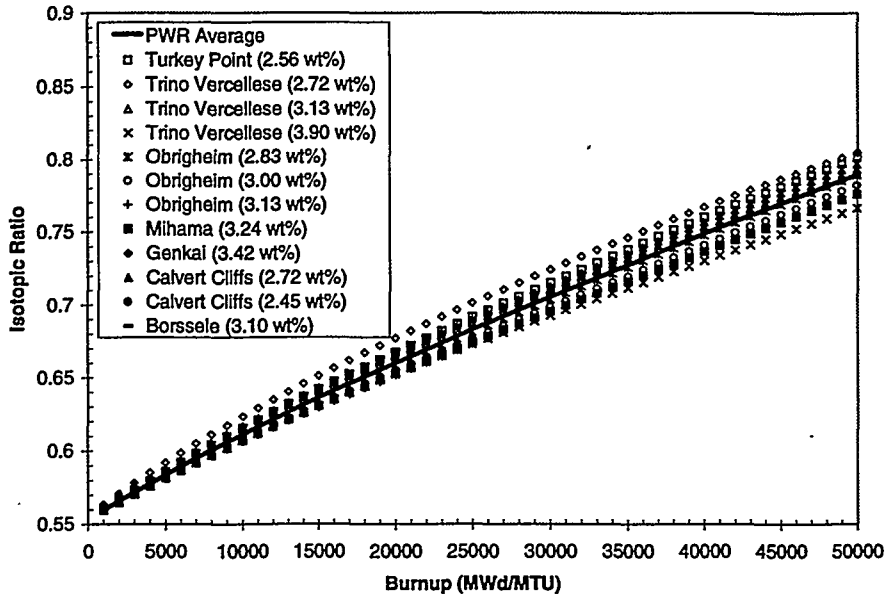

Fig. 20. $^{132}Xe/^{134}Xe$ isotopic ratio versus burnup for several PWRs and for average PWR.

Figures 21-26 contain plots of the calculated xenon and krypton isotopic ratios versus burnup (except for $^{85}Kr/^{86}Kr$ and $^{136}Xe/^{134}Xe$) for several of the reactor types included in the Inverse Problem database. As can be seen each of the isotopic ratios varies differently for each reactor type, and the spread of the isotopic ratios can at times be very large (e.g., $^{130}Xe/^{134}Xe$). It is interesting to note that the $^{131}Xe/^{134}Xe$ isotopic ratio is significantly less dependent upon reactor type than the other xenon isotopic ratios. Also, the $^{131}Xe/^{134}Xe$ and $^{83}Kr/^{86}Kr$ isotopic ratios are not monotonic. Thus, it is apparent that at least two isotopic ratios must be used to determine any information of interest about a reprocessed fuel even in the case of the Forward Problem.

It is also interesting to note that the PWR and BWR isotopic ratio functions are very similar; however, there is a significant difference between the PWR and BWR

functions for the $^{130}Xe/^{134}Xe$ isotopic ratio. From the other ratios alone it is difficult to distinguish between PWR and BWR reactors. In this particular case, it may have been more appropriate to define a single reactor type as light water reactor (LWR). This becomes more apparent when we consider the fact that from a proliferation standpoint there is not a significant difference between PWR and BWR power reactors. For the purposes of this project, we retained the distinction between PWR and BWR reactors and will simply inform the users of the technique that it is highly probable that fissiogenic gas from PWR fuel could easily be confused with fissiogenic gas from BWR fuel and vice versa.

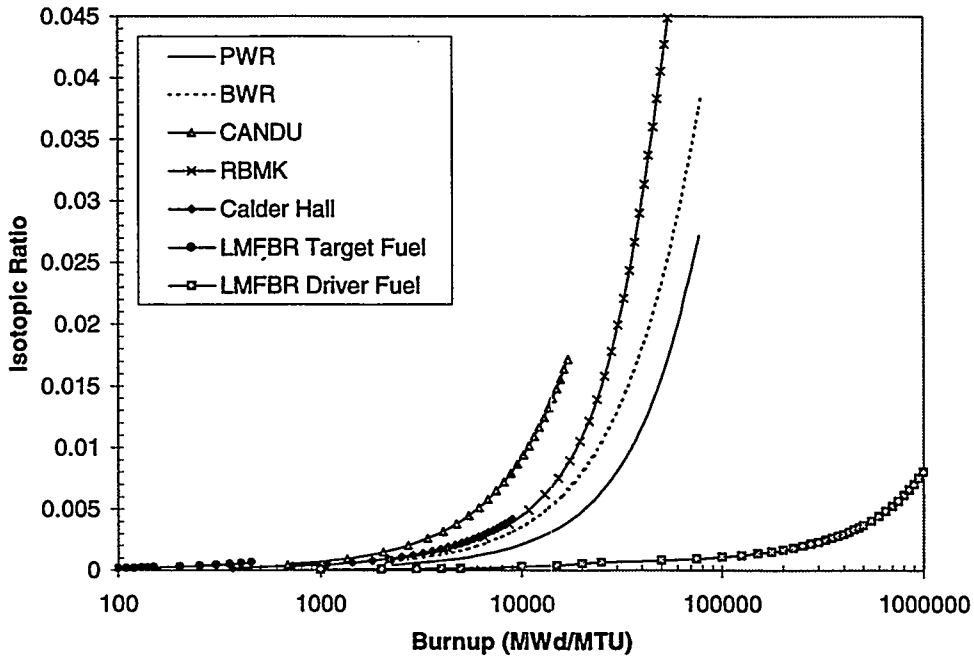

Fig. 21. $^{130}Xe/^{134}Xe$ isotopic ratio versus burnup for all reactor models.

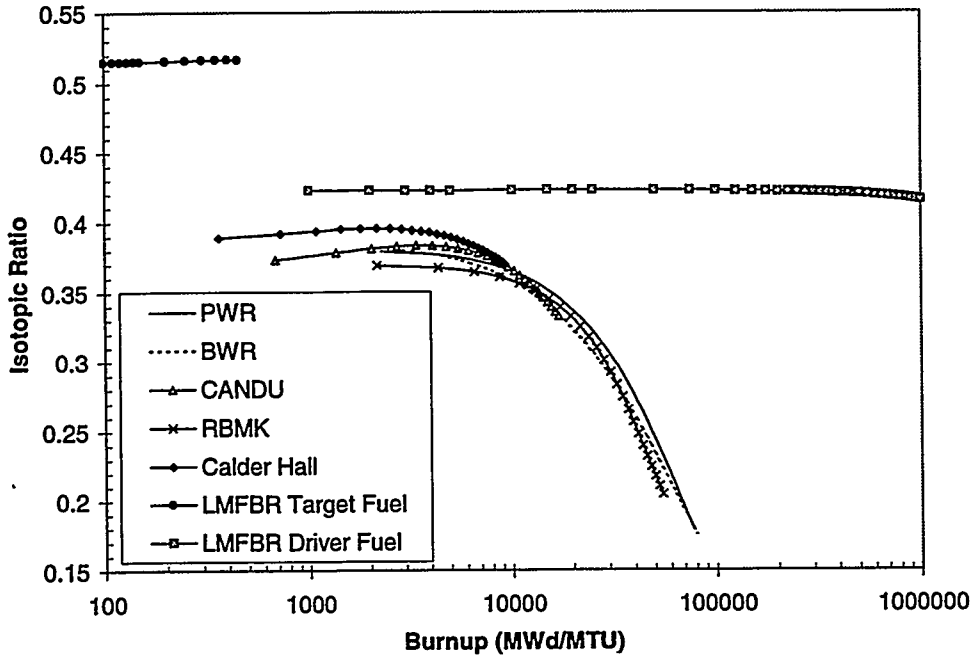

Fig. 22. $^{131}Xe/^{134}Xe$ isotopic ratio versus burnup for all reactor models.

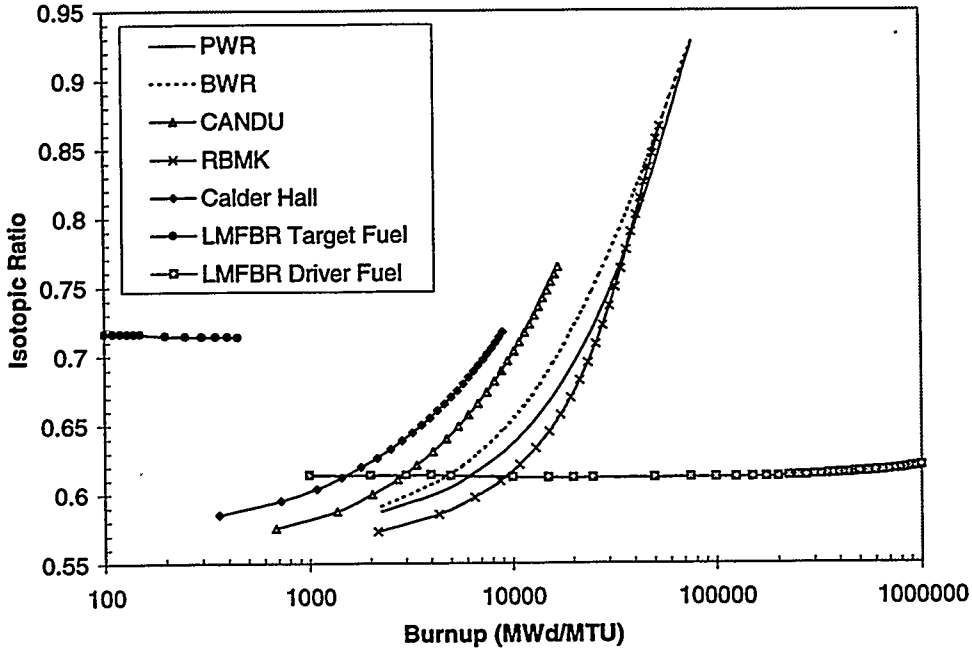

Fig. 23. $^{132}Xe/^{134}Xe$ isotopic ratio versus burnup for all reactor models.

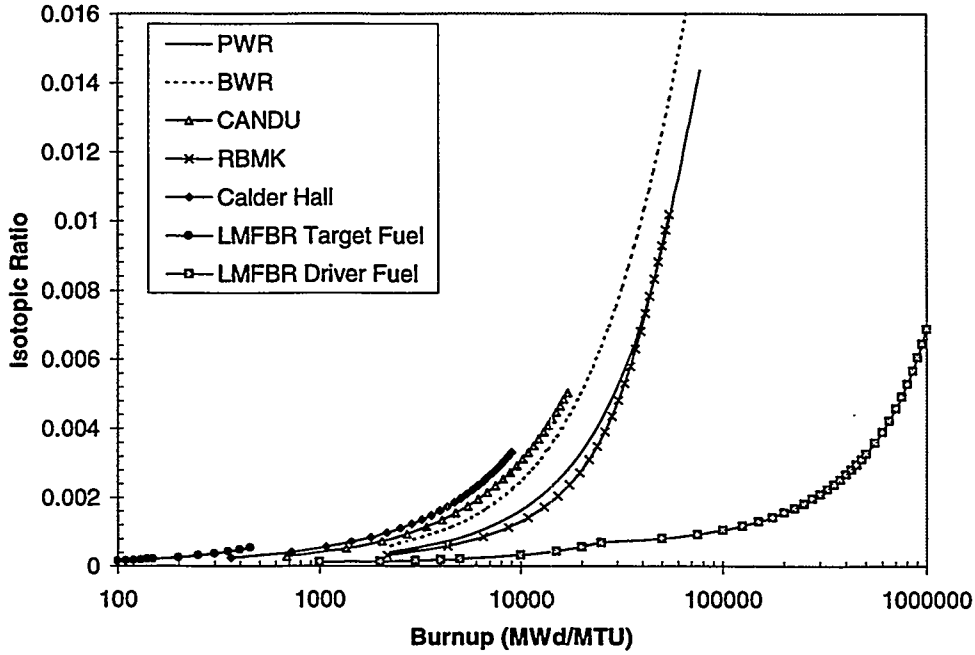

Fig. 24. $^{82}Kr/^{86}Kr$ isotopic ratio versus burnup for all reactor models.

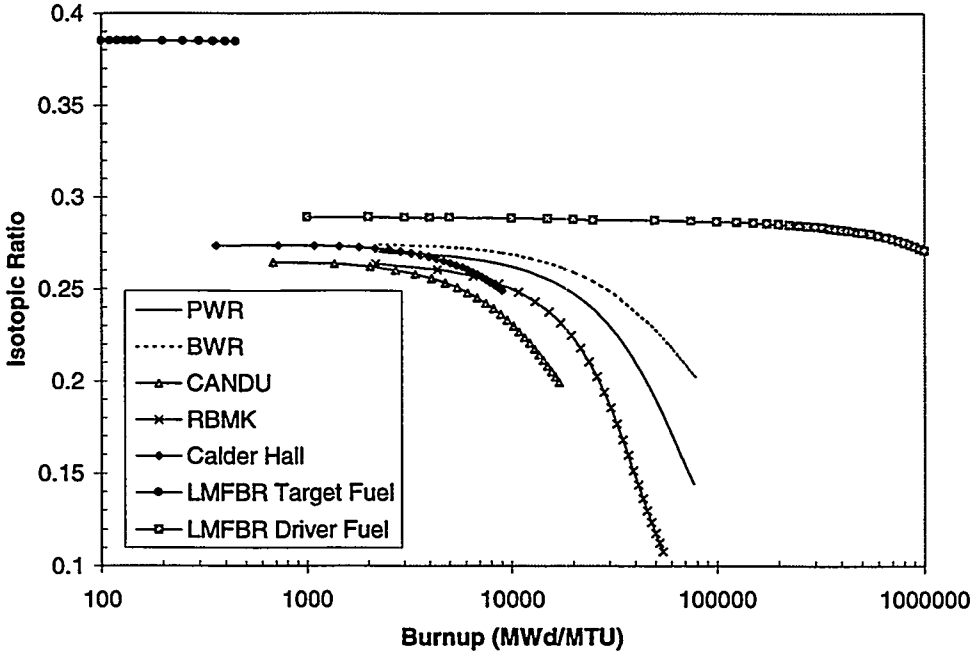

Fig. 25. $^{83}Kr/^{86}Kr$ isotopic ratio versus burnup for all reactor models.

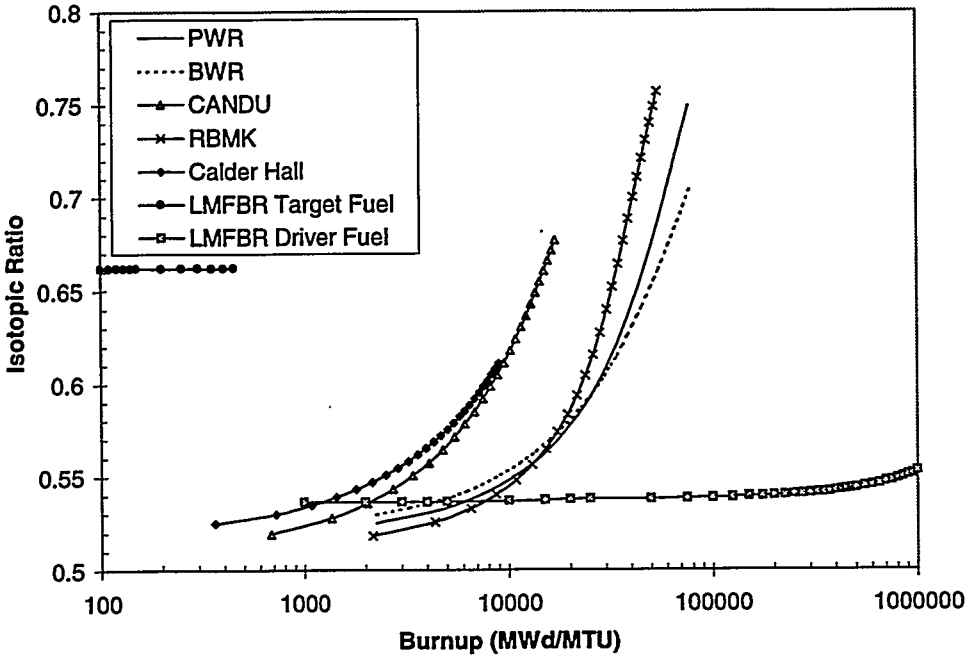

Fig. 26. $^{84}Kr/^{86}Kr$ isotopic ratio versus burnup for all reactor models.

The graphite-moderated reactor results (e.g., Calder Hall and RBMK) in the databases have not been thoroughly tested; however, since Monteburns 3.01 uses detailed physics in its analyses, it is expected that these isotopic ratios will be sufficiently accurate for the environmental monitoring application. On the other hand, the high temperature gas reactor (HTGR) may not have been well simulated using the methodology described above. HTGRs consist of small coated particle fuels mixed into a pin with a carbonaceous binder. These pins are then assembled into a graphite block, which serves as the moderator. This geometry may lead to a double heterogeneity effect (i.e., one in the pin and another in the particle fuel). This effect has not been addressed in the HTGR simulations used in creating the databases. Studying

this effect in more detail is beyond the scope of this project; however, the user should be aware of this incompleteness. In the event of the reprocessing of HTGR fuel, the benchmarking of the reactor physics database should be extended to include this fuel type.

The databases that were created contain numerous nuclides. Monteburns' ability to predict the concentrations of the non-noble gas nuclides in spent fuel has not been extensively benchmarked; however, Monteburns ability to predict the $^{240}Pu/^{239}Pu$ isotopic ratio was examined using the experimental data reported above (Table X). It was determined that Monteburns could consistently calculate the $^{240}Pu/^{239}Pu$ isotopic ratio for numerous fuel types to within ±0.9%. This degree of accuracy is similar to that found for other reactor code systems; however, if this information is to be used more extensively in the future, the Monteburns code system should be benchmarked more rigorously.

V. DATA ANALYSIS METHODOLOGY

The measured isotopic ratios (generated using the system described in Section III) were coupled to the calculated fissiogenic gas database (described in Section IV) through a Bayesian analysis technique which allowed for the determination of the most likely fuel type and burnup from a set of measured isotopic ratios. A brief description of Bayesian analysis theory and development, along with the procedure used in these analyses, is given in this section. Also, the methodology used for removing the background air contaminant is given.

A. Background Air Removal

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. Since some noble gas isotopes (for instance, ^{129}Xe and ^{80}Kr) 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 the xenon and krypton isotopes in air (either assumed or measured).

The on-stack sample will contain both fissiogenic gases and natural-air gases. Thus, the measured concentration will be given by

$$N^x_{m,u} = N^x_{f,u} + N^x_{m,air} \tag{10}$$

where $N^{x}_{f,u}$ is the fissiogenic component of the concentration of isotope x in the unknown sample, $N^{x}_{m,u}$ is the measured concentration of fissiogenic isotope x in the unknown sample, and $N^{x}_{m,air}$ is the concentration of fissiogenic isotope x in atmospheric-air.

If we assume that the isotope ^{129}Xe has no fissiogenic component, then the concentration of ^{129}Xe in the unknown sample is given by

$$N^{129}_{m,u} = N^{129}_{m,air} \tag{11}$$

and the ratio of the measured fissiogenic isotope x concentration on-stack to that measured for ^{129}Xe will be given by

$$\frac{N^{x}_{m,u}}{N^{129}_{m,u}} = \frac{N^{x}_{f,u}}{N^{129}_{m,air}} + \frac{N^{x}_{m,air}}{N^{129}_{m,air}} \tag{12}$$

where $N^{129}{}_{m,air}$ is the concentration of the non-fissiogenic isotope (in this case ^{129}Xe) in atmospheric-air and $N^{129}{}_{m,u}$ is the measured concentration of the non-fissiogenic isotope in the unknown sample.

If the abundances of all of the measured noble gas isotopes in atmospheric air (relative to the abundance of the non-fissiogenic isotope) are known, then the fissiogenic component of that isotope can be determined by solving Eq. (11) for the ratio of the fissiogenic component of the sample or

$$\frac{N^{x}_{f,u}}{N^{129}_{m,air}} = \frac{N^{x}_{m,u}}{N^{129}_{m,u}} - \frac{N^{x}_{m,air}}{N^{129}_{m,air}}. \tag{13}$$

Considering the element xenon with normalizing isotope ^{134}Xe and non-fissiogenic isotope ^{129}Xe, the isotopic ratio of interest is given by

$$\frac{N^{x}_{f,u}}{N^{134}_{f,u}} = \frac{\left(\frac{N^{x}_{m,u}}{N^{129}_{m,u}} - \frac{N^{x}_{m,air}}{N^{129}_{m,air}}\right)}{\left(\frac{N^{134}_{m,u}}{N^{129}_{m,u}} - \frac{N^{134}_{m,air}}{N^{129}_{m,air}}\right)}. \tag{14}$$

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. A similar technique can be used for the krypton gases using ^{78}Kr and/or ^{80}Kr as the gas lacking a significant fissiogenic component.

B. Bayesian Analysis Theory

The Bayesian analysis technique is used to aid in determining the most likely hypothesis or event that would lead to any set of observed data.[72] For any observed phenomena there may be numerous events that could have led to that observation (Fig. 27). For instance, the observed phenomena may be that an accident occurred (e.g., a bridge collapsed), and the set of events could be different potential scenarios that led to that accident (e.g., a weight limit was exceeded or a vibrational resonance was created in the structure). Using probability estimations, Bayesian analysis can aid in determining which scenario was the most likely to have led to the accident.

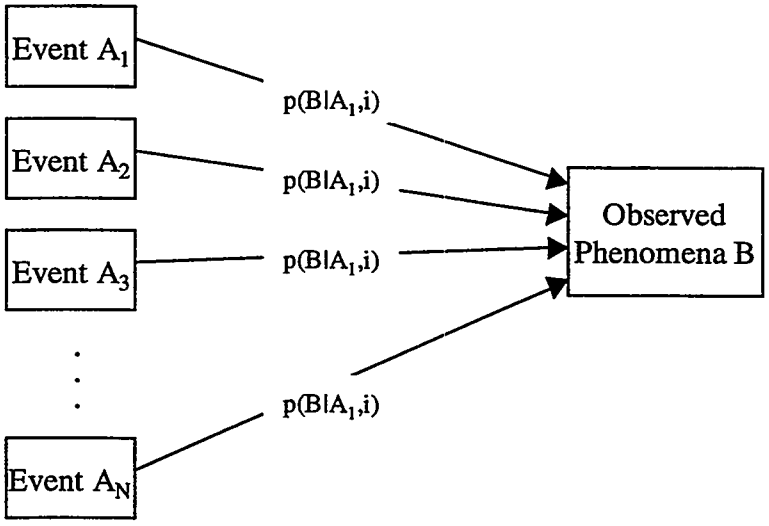

Fig. 27. Generation of observed phenomena via multiple potential events.

In all the probability discussions below, the standard probability notation has been used. In this case, the notation $p(B|A_I I)$ is read as the probability that one would observe B given that A_I occurred and information I is true. This notation is based on the fundamentals of Boolean algebra.[73]

In the environmental monitoring technique developed here, our interest is in determining which of a set of proposed reactor models (M) at any of a number of possible burnups (B) is most likely to have led to the measurement of a set of isotopic ratios (R) accounting for any potential background information (I) available. The analyses used here will be based on two fundamental rules of probability:

$$p(AB \mid I) = p(A \mid I)\, p(B \mid AI) \tag{15}$$

$$p(A \mid I) + p(\overline{A} \mid I) = 1, \tag{16}$$

where AB represents the logical product of propositions A and B and $\overline{A}$ represents the logical complement of A. These two rules are called the product and sum rules, respectively.[74] Since Boolean algebra is commutative, it can be shown that $p(AB|I) = p(BA|I)$. Using this relationship and the product rule, the fundamental theorem of Bayesian analysis can be derived as follows:

$$p(AB \mid I) = p(BA \mid I), \tag{17}$$

$$p(A \mid I)p(B \mid AI) = p(B \mid I)p(A \mid BI), \tag{18}$$

$$p(B \mid AI) = \frac{p(B \mid I)p(A \mid BI)}{p(A \mid I)}. \tag{19}$$

Equation (19) is called Bayes' theroem,[75] and it relates the probability that the hypothesis (B) is true, given the data (A), to the probability that we would observe the measured data (A) if the hypothesis (B) were true. In simplest terms, Bayes' theorem can be viewed as

$$prob(hypothesis|data, I) \propto prob(data|hypothesis, I) \cdot prob(hypothesis|I). \tag{20}$$

In order to use Bayes' theorem effectively, the denominator in Eq. (19) must be expressed in terms, which can be evaluated in practice. This will require the development of two rules derived directly from the product and sum rules.

It can be shown that

$$\overline{A} + \overline{B} = \overline{AB} \tag{21}$$

and similarly

$$A + B = \overline{\overline{A}\,\overline{B}}. \tag{22}$$

Using the sum rule, we can show that

$$p(A+B\,|\,I)+p(\overline{A+B}\,|\,I)=1 \tag{23}$$

$$p(A+B\,|\,I)+p(\overline{\overline{A}\,\overline{B}}\,|\,I)=1 \tag{24}$$

$$p(A+B\,|\,I)=1-p(\overline{A}\,\overline{B}\,|\,I) \tag{25}$$

Using the product rule and the sum rule, the following can be acquired:

$$p(A+B\,|\,I)=1-p(\overline{A}\,|\,\overline{B}I)p(\overline{B}\,|\,I) \tag{26}$$

$$p(A+B\,|\,I)=1-\left[1-p(A\,|\,\overline{B}I)\right]p(\overline{B}\,|\,I) \tag{27}$$

$$p(A+B\,|\,I)=1-p(\overline{B}\,|\,I)+p(A\,|\,\overline{B}I)p(\overline{B}\,|\,I) \tag{28}$$

$$p(A+B\,|\,I)=1-p(\overline{B}\,|\,I)+p(A\overline{B}\,|\,I)\,. \tag{29}$$

Using the sum rule again, it can be shown that

$$p(A+B\,|\,I)=p(B\,|\,I)+p(A\overline{B}\,|\,I)\,. \tag{30}$$

Using the product rule again, yields

$$p(A+B\,|\,I)=p(B\,|\,I)+p(\overline{B}\,|\,AI)p(A\,|\,I)\,. \tag{31}$$

Finally, if the sum rule and product rule are applied once more, we acquire

$$p(A+B\,|\,I)=p(B\,|\,I)+\left[1-p(B\,|\,AI)\right]p(A\,|\,I) \tag{32}$$

$$p(A+B\,|\,I)=p(B\,|\,I)+p(A\,|\,I)-p(A\,|\,I)p(B\,|\,AI) \tag{33}$$

$$p(A+B\,|\,I)=p(A\,|\,I)+p(B\,|\,I)-p(AB\,|\,I)\,. \tag{34}$$

Equation (34) is commonly termed the Generalized Sum Rule.[76] It can be shown that if A and B are mutually exclusive, then $p(AB|I)=0$ and

$$p(A+B\,|\,I)=p(A\,|\,I)+p(B\,|\,I)\,. \tag{35}$$

If [A=(A_1, A_2, A_3, ... A_n)] is a set of exhaustive, mutually exclusive propositions, and B is an arbitrary proposition (B∈A), then it can be shown that

$$p(B \mid I) = p(BA_i \mid I) \quad where \quad A_i = B \tag{36}$$

and

$$p(BA_i \mid I) = 0 \quad if \quad A_i \neq B\,. \tag{37}$$

Thus, from the sum rule, we acquire

$$p(B \mid I) = \sum_{i=1}^{n} p(BA_i \mid I)\,. \tag{38}$$

Using the product rule, Eq. (38) becomes

$$p(B \mid I) = \sum_{i=1}^{n} p(B \mid A_i I)\, p(A_i \mid I)\,. \tag{39}$$

Equation (39) is often termed "marginalization".[76] Substituting Eq. (39) into Bayes' theorem [Eq. (19)], we acquire

$$p(A_i \mid BI) = \frac{p(B \mid A_i I)\, p(A_i \mid I)}{\sum_{i=1}^{n} p(B \mid A_i I)\, p(A_i \mid I)}\,. \tag{40}$$

Equation (40) can now be used to evaluate the probability that the proposition A_i is correct given the data B and the information I.

To use Eq. (40), the probabilities must be evaluated. It has been shown that the most conservative distribution for these probabilities is the Gaussian distribution,[77]

$$p(x \mid I) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{(x-\mu)^2}{2\sigma^2}\right], \tag{41}$$

where μ and σ are real parameters describing the mean and standard deviations of the distribution. This is called assigning probabilities using the Maximum Entropy Formulation, since the Gaussian distribution can be derived from the "information entropy" of any distribution.[77] Thus using Eqs. (40) and (41), we can determine the probability that any proposition (B) is true given a mutually exclusive and exhaustive set of propositions (A), any background information (I), and knowledge of the distributions describing the proposition set (A).

C. Analysis Procedure

Given a set of I measured isotopic ratios [R^m=(R_1^m, R_2^m, ...R_I^m)], their associated standard deviations (σ_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 the Bayesian analysis methodology described above. 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 (σ_{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 first step in this analysis is to determine a burnup value for each measured isotopic ratio for each model (B_{ij}^{int}). This was accomplished by linearly interpolating within the database. Thus assuming that R_i^m is between R_{ijk}^c and R_{ijk+1}^c then an interpolated burnup value can be determined as follows:

$$B_{ij}^{\text{int}} = B_{jk}^c + \frac{(R_i^m - R_{ijk}^c)(B_{jk+1}^c - B_{jk}^c)}{(R_{ijk+1}^c - R_{ijk}^c)}. \qquad (42)$$

The standard deviation of this interpolated burnup value can be calculated using the following expressions:

$$\sigma_{ij}^{\text{int}+} = B_{jk+1}^{c} - B_{ij}^{\text{int}} + \frac{(R_i^m + \sigma_i^m - R_{ijk+1}^c + \sigma_{ijk+1}^c)(B_{jk+1}^c - B_{jk}^c)}{(R_{ijk+1}^c - \sigma_{ijk+1}^c - R_{ijk}^c + \sigma_{ijk}^c)} \tag{43}$$

$$\sigma_{ij}^{\text{int}-} = B_{ij}^{\text{int}} - B_{jk}^{c} - \frac{(R_i^m - \sigma_i^m - R_{ijk}^c - \sigma_{ijk}^c)(B_{jk+1}^c - B_{jk}^c)}{(R_{ijk+1}^c + \sigma_{ijk+1}^c - R_{ijk}^c - \sigma_{ijk}^c)} \tag{44}$$

$$\sigma_{ij}^{\text{int}} = \frac{(\sigma_{ij}^{\text{int}+} + \sigma_{ij}^{\text{int}-})}{2} \tag{45}$$

Using these interpolated values for each measured isotopic ratio, a combined average burnup (B_j) and an associated standard deviation (σ_j) for each model (M_j) can be determined using

$$B_j = \frac{\sum_i B_{ij}^{\text{int}} / (\sigma_{ij}^{\text{int}})^2}{\sum_i 1 / (\sigma_{ij}^{\text{int}})^2} \tag{46}$$

$$\sigma_j = \left[\sum_i 1 / (\sigma_{ij}^{\text{int}})^2 \right]^{-1/2}. \tag{47}$$

Using this combined average burnup, we can interpolate again in the database to generate calculated isotopic ratios (R_{ij}^{μ}), and their standard deviations (σ_{ij}^{μ}), that correspond to the average burnup (B_j) for each model (M_j). Given that the average burnup (B_j) is between two consecutive points in the database (B_{jk}^{c} and B_{jk+1}^{c}) then

$$R_{ij}^{\mu} = R_{jk}^{c} + \frac{(B_j - B_{jk}^c)(R_{ijk+1}^c - R_{ijk}^c)}{(B_{jk+1}^c - B_{jk}^c)}. \tag{48}$$

The standard deviation of this interpolated burnup value can be determined using the following expressions:

$$\sigma_{ij}^{\mu+} = \sigma_{ijk}^{c} + R_{ijk}^{c} - R_{ij}^{\mu} + \frac{(B_j + \sigma_j - B_{jk}^{c})(R_{ijk+1}^{c} + \sigma_{ijk+1}^{c} - R_{ijk}^{c} - \sigma_{ijk}^{c})}{(B_{jk+1}^{c} - B_{jk}^{c})} \quad (49)$$

$$\sigma_{ij}^{\mu-} = \sigma_{ijk}^{c} - R_{ijk}^{c} + R_{ij}^{\mu} - \frac{(B_j - \sigma_j - B_{jk}^{c})(R_{ijk+1}^{c} - \sigma_{ijk+1}^{c} - R_{ijk}^{c} + \sigma_{ijk}^{c})}{(B_{jk+1}^{c} - B_{jk}^{c})} \quad (50)$$

$$\sigma_{ij}^{\mu} = \frac{(\sigma_{ij}^{\mu+} + \sigma_{ij}^{\mu-})}{2}. \quad (51)$$

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 the be calculated using the maximum entropy formulation:

$$p(R_i^m \mid M_j, B_j, E) = \frac{1}{\sigma_{ij}^{\mu}\sqrt{2\pi}} \exp\left[-\frac{(R_i^m - R_{ij}^{\mu})^2}{2(\sigma_{ij}^{\mu})^2}\right]. \quad (52)$$

Using these model based probabilities and Bayes' theorem [Eq. (40)], 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

$$p(M_j, B_j \mid R^m, E) = \frac{p(M_j, B_j \mid E)\prod_{i=1}^{I} p(R_i^m \mid M_j, B_j, E)}{\sum_{j=1}^{J}\prod_{i=1}^{I} p(R_i^m \mid M_j, B_j, E)\, p(M_j, B_j \mid E)}. \quad (53)$$

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. This background

evidence could be information such as an inspector's observation that the fuel is not of a specific type, knowledge that a country does not possess a certain type of reactor, or any other data to which an inspector might have access. If there is no reason to prefer one model over any of the others then the priors can all be set to unity.

It should be noted that the analysis described above is primarily for use in the Inverse Problem and has assumed that our database includes a mutually exclusive and exhaustive set of reactor models. Since the database used here contains fourteen different fuel types each evaluated over an extensive range of burnups, it will be assumed that this assumption is valid. However, one should note that if a reactor fuel of some type markedly different than those included here is reprocessed, this assumption will no longer be appropriate and other methods should be used (or the database should be modified).

For the Forward Problem the analysis is greatly simplified. In this case, assuming that the fuel type is already known, the problem is simply to determine the probability that the measured isotopic ratios from the given fuel at a specific burnup are observed. This analysis is simply given by

$$p(R^m \mid M_j, B_j, E) = \prod_{i=1}^{I} p(R_i^m \mid M_j, B_j, E) \tag{54}$$

where B_j is determined using the same procedure as described previously (i.e., interpolation and a combined average) and the probabilities are determined from Eq. (52). In this case, no comparisons to other models are necessary.

The methodology for determining the fuel age is also straightforward. Given a spent fuel type and burnup, the value of the $^{85}Kr/^{86}Kr$ isotopic ratio at discharge can be predicted. Thus, given a measurement of the same ratio, the fuel age can be determined as follows:

$$t = -\frac{1}{\lambda}\ln\left(\frac{R(t)}{R_0}\right) \tag{55}$$

where t is the fuel age, λ is the decay constant of ^{85}Kr, $R(t)$ is the isotopic ratio at time t, and R_0 is the isotopic ratio at discharge. Equation (55) is derived directly from the simple exponential decay equation and assumes that the decay of all parent isotopes to ^{85}Kr is instantaneous.

The procedure above was coded for use in developing the environmental monitoring technique. The actual code will be described in greater detail later (Section VI).

VI. COMPLETED MONITORING SYSTEM

All of the pieces essential to constructing the monitoring technique (i.e., the measurement system, reactor physics databases, and data analysis technique) have been developed in the previous sections. This section will describe in detail the integration of these portions and the user interface developed for use with the environmental monitoring technique. Also, included below is a description of the validation of the integrated system using on-stack samples and using literature experimental data.

A. Description of Monitoring System

The complete monitoring technique has been developed as described in Section II. A Visual Basic code was written that incorporates the calculated reactor physics databases (Section IV.C) and the Bayesian analysis procedure (Section V.C.). This code was generated to be used as a user-friendly tool for applying this environmental monitoring technique. The code was named NOVA (NOble gas enVironmental monitoring Application). This code allows the user to input any number of measured isotopic ratios (background corrected or not background corrected), performs the correction for background air (if necessary), analyzes the ratios for either the Inverse or Forward Problem as specified by the user, and outputs the solution including expected plutonium 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. Each portion of the code is described in greater detail below.

NOVA consists of eight "forms". Each form is an interactive page that can be used to perform functions, input information, or display solutions. The first form in NOVA is simply the "Noble Gas Program" page (Fig. 28). This page displays the code title and has three buttons at the bottom. Clicking the "Authors" button displays the authors page (Fig. 29) which simply displays the names of all individuals contributing to the development of the NOVA code and the environmental monitoring methodology. This page also includes the address of the primary author who can be contacted for technical support or additional information concerning the code.

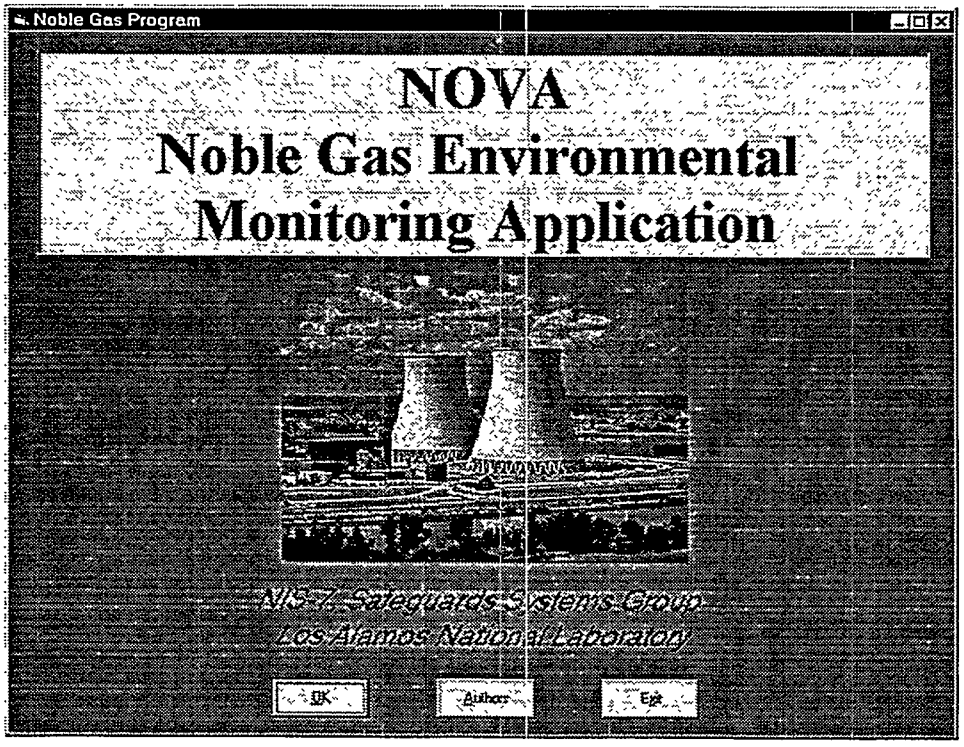

Fig. 28. The "Noble Gas Program" form in NOVA.

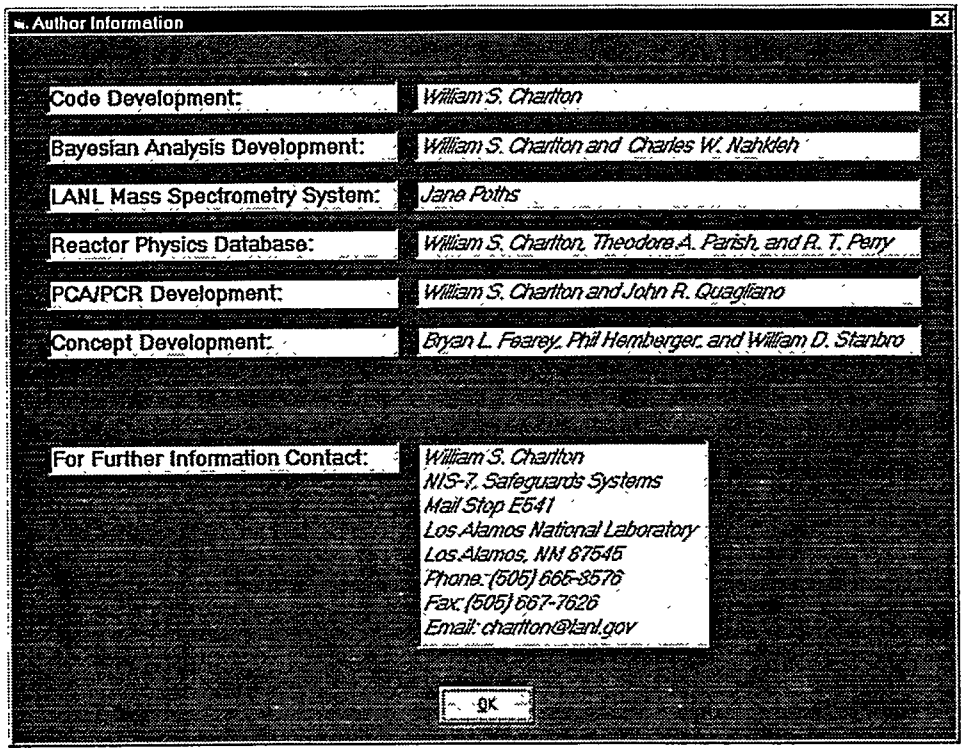

Fig. 29. The "Authors Information" form in NOVA.

The "Exit" button on the "Title" form (Fig. 28) allows the user to exit the program. The "OK" button on the "Noble Gas Program" form brings up the "Input Data" form (Fig. 30). The "Input Data" form is used to input any measured data for use in the analysis. This form has places for entering any of a number of isotopic ratios (all normalized to ^{129}Xe or ^{80}Kr). Note that for proper execution, the user must enter at least two isotopic ratios (not including the $^{136}Xe/^{129}Xe$ or $^{85}Kr/^{80}Kr$, which are not used for burnup and fuel type determination) and one of the normalizer ratios (i.e., $^{134}Xe/^{129}Xe$ or $^{86}Kr/^{80}Kr$). The "Input Data" form also has space allotted for entering the measured isotopic ratios for natural air or if the user desires a set of default values

for the xenon and krypton natural air isotopic ratios is available. Also, if the data being input has already been background corrected (or did not require a background correction) then checking the "Eliminate background air correction" checkbox will enter all zeros for the background air values. Three buttons are located at the bottom of the "Input Data" form. The last button ("Cancel") simply returns the user to the "Noble Gas Program" form. The "Advanced" button will open a new form called the "Advanced" form (Fig. 31) which allows the user to select various options for the analysis. The "Analyze" button will open the "Data Confirmation" form (Fig. 32) which allows the user to confirm the data entered prior to analysis.

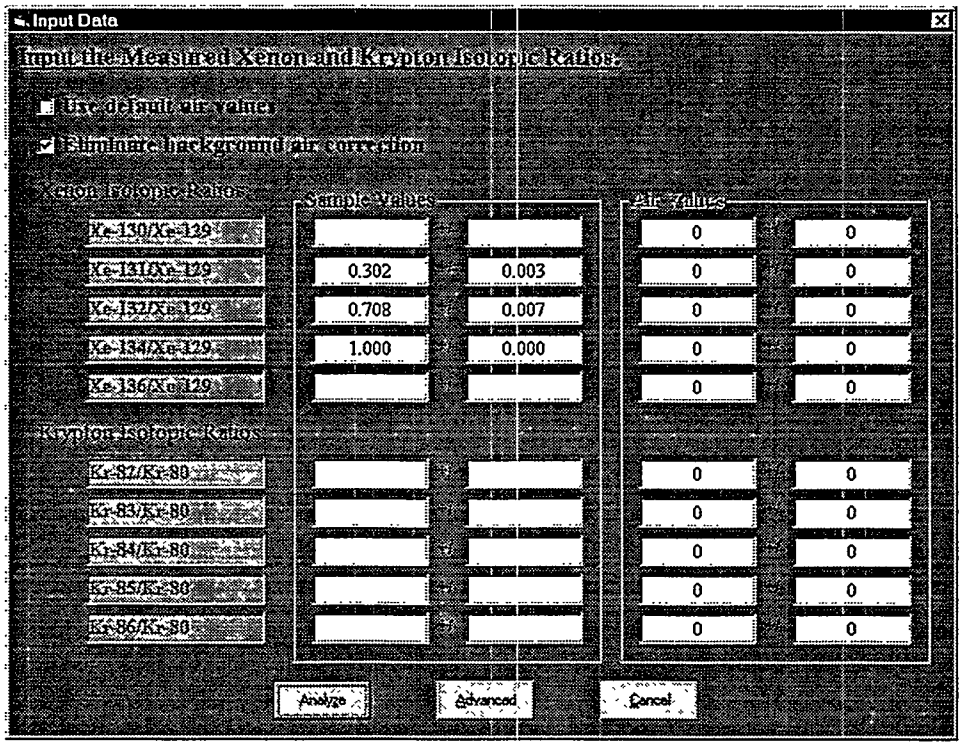

Fig. 30. The "Input Data" form in NOVA.

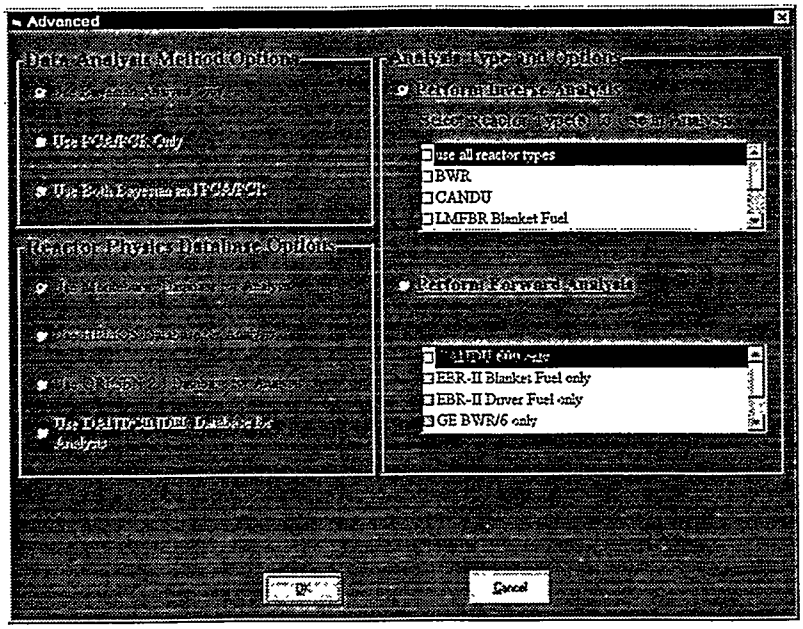

Fig. 31. The "Advanced" form in NOVA.

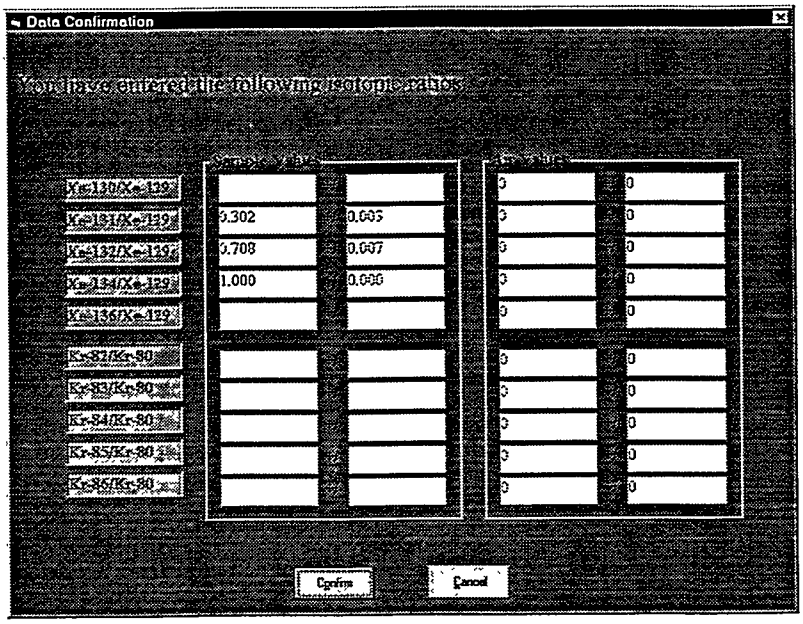

Fig. 32. The "Data Confirmation" form in NOVA.

The "Advanced" form gives the user several options for performing the analysis. The first option allows selection of the method used to perform the data analysis. The only option currently available here is the Bayesian analysis option. The other options are reserved for future developments of the code. The second option available allows the user to choose which reactor physics database to use. The default database is the Monteburns database; however, several other databases are available if the user wishes to test the other available code systems. The last option available is to select the Forward Problem or the Inverse Problem. The default option for the code is the Inverse Problem. In addition, the user can use the listbox available for selecting and eliminating certain reactor types from the analysis. The user can also select to perform a Forward Problem analysis in which case the known fuel type must be selected from the listbox.

The "Data Confirmation" form is used simply to allow the user to double check the input values prior to performing the analysis. The data entered on the "Input Data" form is displayed here. By pressing the "Cancel" button, the user is taken back to the "Input Data" form so that the input values can be corrected. Pressing the "Confirm" button will execute the analysis. While the analysis is being executed the "Analysis Progress" form (Fig. 33) is displayed which simply shows the progress of the analysis. The progress is represented by a progress bar. When this bar reaches 100% the calculation is complete.

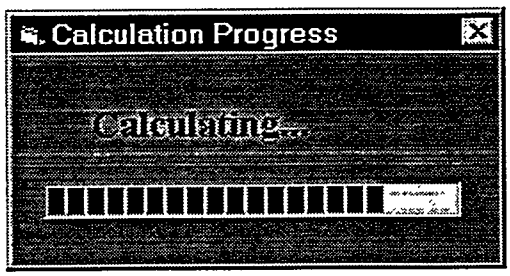

Fig. 33. The "Calculation Progress" form in NOVA.

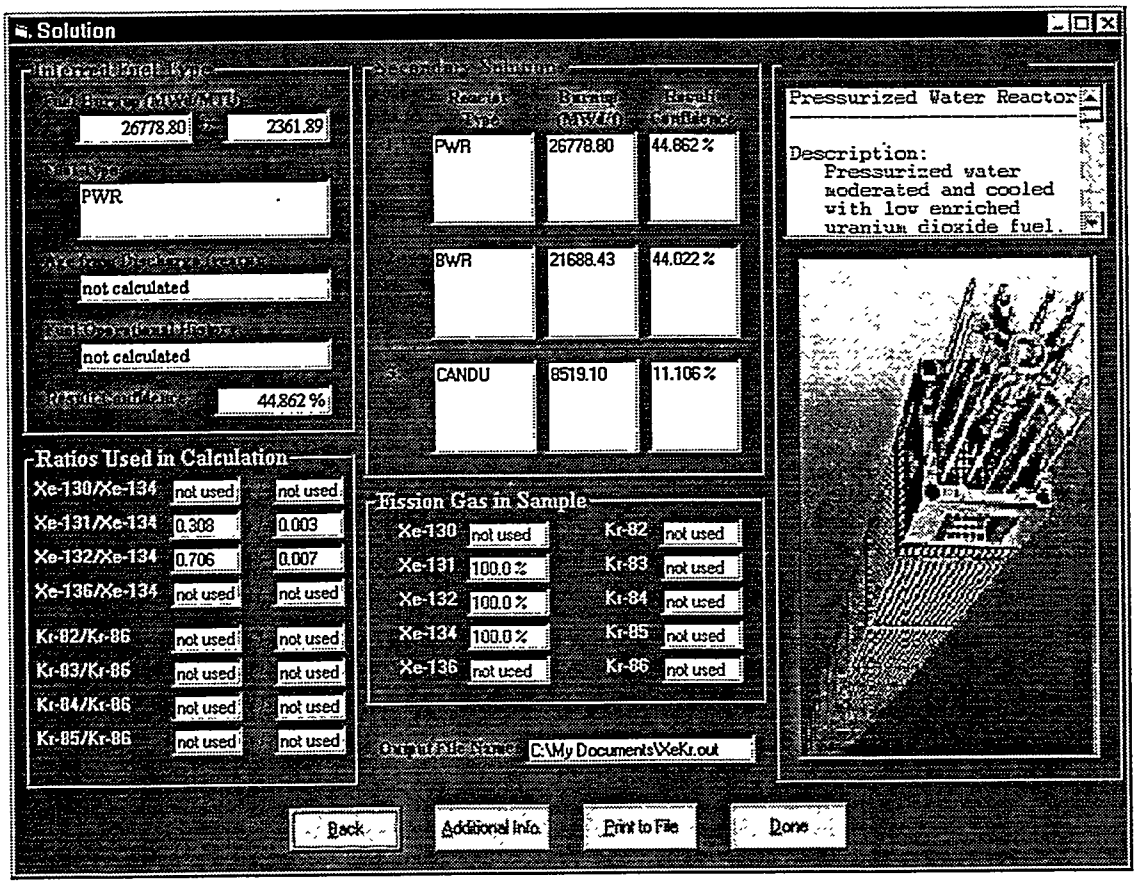

Fig. 34. The "Solution" form in NOVA.

When the analysis is complete, the "Solution" form is displayed (Fig. 34). The "Solution" form displays various results calculated during the analysis including the most likely burnup and fuel type for the input isotopic ratios, the fuel age, and the result confidence for this solution. This page also displays the secondary solutions (in order of likeliness), the percent fission gas in the sample, and a short description (including a picture) of the inferred fuel type. Several buttons are located on this page. The "Print to File" button will print the solution information to the file specified in the "Output File Name" textbox (which can be altered as the user sees fit). The "Back" button will return the user to the "Input Data" form. The "Done" button will return the user to the "Noble Gas Program" form.

The last button on the "Solution" form (the "Additional Info." button) will display the "Additional Information" form. This form displays all manner of information pertaining to the most likely solution. Plutonium concentrations, fission product concentrations, and minor actinide concentrations are listed, as well as any comments pertaining to this information. Also, the calculated and input isotopic ratios are displayed for comparison. This information could also be used to aid in removing any statistical outliers in the input data. The "Back" button on the "Additional Information" form is used to return the user to the "Solution" form.

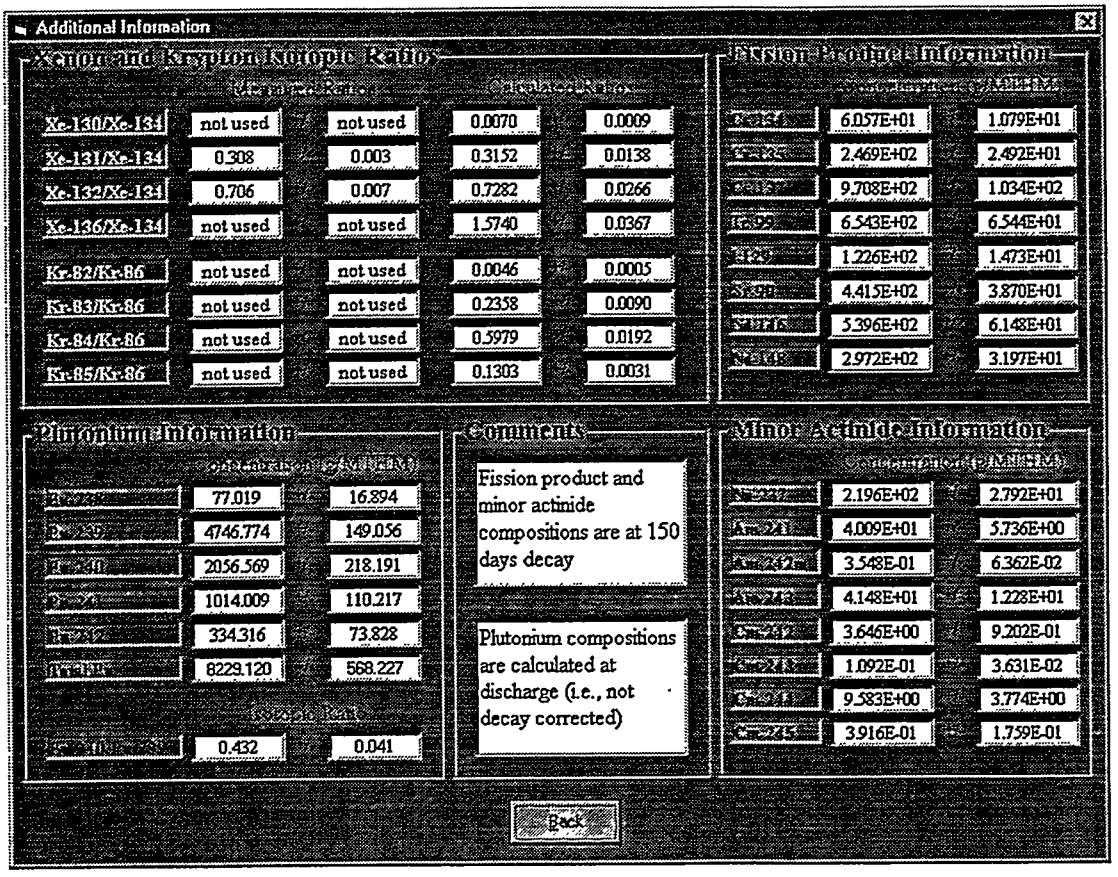

Fig. 35. The "Additional Information" form in NOVA.

The NOVA code was designed to be as user-friendly as possible while still displaying a vast amount of information. The usage of this code is straightforward and requires minimal training. A strategy for maximizing the effective use of NOVA could include selectively applying the Forward and Inverse Problem options and searching the other reactor physics databases (especially if the user feels that one reactor physics code may be more accurate for a specific reactor type). Other strategies will be left to

the user to explore. The actual coding for each of the forms used in the NOVA code is listed in Appendix C.

B. System Validation and Benchmarking

The NOVA code and the LANL mass spectrometry system was used to analyze several sets of air samples to determine the validity of the proposed system for determining spent fuel burnup and fuel type. The NOVA code was also tested on available experimental data from the literature to confirm its effectiveness for as many fuel types as possible. Also included below is a description of the usefulness of the $^{136}Xe/^{134}Xe$ isotopic ratio for confirming declared power histories.

1. U.S. Production Reactor Target Fuel

The proposed environmental monitoring technique (and the NOVA code) have been validated using on-stack measurements taken during the reprocessing of spent target fuel from a U.S. production reactor. The samples were acquired during the dissolution of weapons-grade spent fuel with a declared burnup of 178 MWd/MTU. The complete system generated an inferred burnup of 171 ± 13 MWd/MTU and correctly matched the measured samples to the reactor type (see Figs. 36 and 37). Thus, the inferred burnup was in error by only 3.9%, which is most likely within the uncertainty of the declared burnup.

This fuel represents a good test of the system because of the small signal-to-noise ratio in low-burnup fuel and the increased difficulty in modeling this reactor. On the other hand, the stable noble gas isotopic ratios from the production reactor fuel are

clearly different from those of any other fuel at low burnup values. Therefore, it is unlikely that the analysis would incorrectly determine the reactor type for this fuel dissolution.

This validation demonstrates the accuracy of techniques developed; however, it does not test the ability to distinguish between different power reactor fuels. Below is a test of the PWR and BWR distinguishing characteristics of the NOVA code using experimental data from the literature. Full system tests for power reactor fuels (including on-stack sample collection) will require the cooperation of a commercial reprocessing facility.

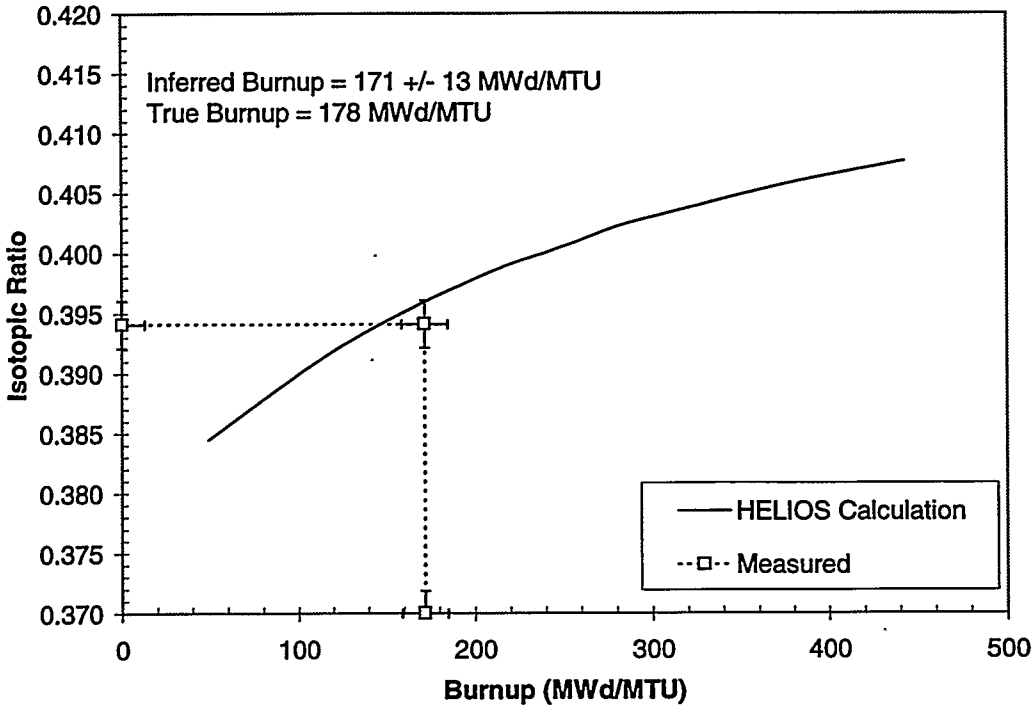

Fig. 36. $^{131}Xe/^{134}Xe$ isotopic ratio versus burnup for U.S. production reactor data.

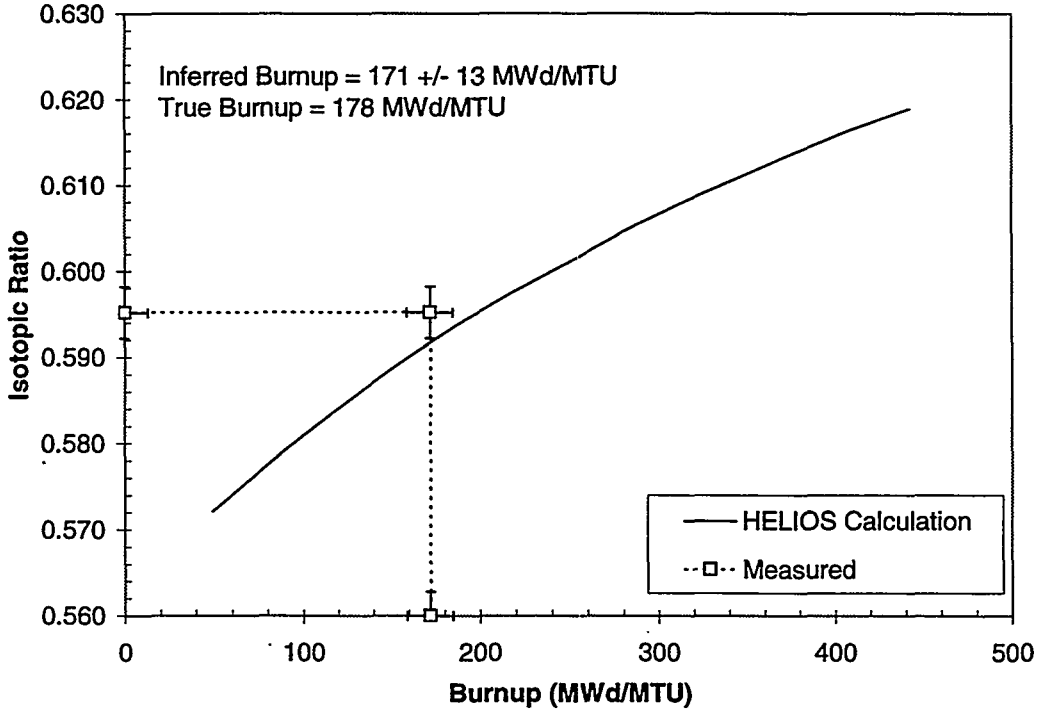

Fig. 37. $^{132}Xe/^{134}Xe$ isotopic ratio versus burnup for U.S. production reactor data.

2. Experimental Breeder Reactor-II Driver and Blanket Fuel

To analyze the proposed technique's ability to distinguish high burnup fuels, air samples were taken from the argon hot-cell at the Idaho National Engineering Laboratory (INEL) following the processing of spent driver fuel from the Experimental Breeder Reactor-II (EBR-II). The fuel processed was of extremely high burnup; however, the operators did not declare a particular burnup value. Also, several fuel samples were processed, so it was unlikely that a single burnup value would be matched directly. The air samples were analyzed using the proposed technique (i.e., mass spectrometry measurement and analysis using NOVA). The analysis resulted in a

prediction of LMFBR driver fuel at a burnup of 268,600 ± 15,800 MWd/MTU. This qualitatively matched the expected value. Again, it was unlikely that the analysis would result in the selection of the wrong fuel type since LMFBR driver fuel is significantly different from any other fuel, but this analysis does help support the versatility of the methodology.

3. Light Water Reactor Fuels.

The most prolific reactor type in existence today is the light water reactor (i.e., PWRs and BWRs). For this reason, it was important that the monitoring technique be capable of accurately predicting the burnup and fuel type for these fuels. It is also interesting to note that the PWR and BWR fuels result in very similar isotopic ratio functions (Fig. 21-29), and it was important to determine if the monitoring technique would be accurate enough to distinguish between these very similar fuel types.

Various measurements in the past have been performed on LWR fuels (see Table X). In order to determine the accuracy and versatility of the monitoring technique for LWRs, the experimental data reported for the Obrigheim[64] (PWR), Borssele[55] (PWR), Gundremmingen[60] (BWR), and Garigliano[59] (BWR) reactors was analyzed using the NOVA code (using the Inverse Problem option). These fuels were chosen because of their numerous measurements and also because of the increased consistency of their measurements. In general, the measurement uncertainties for these experiments were approximately ±2%. This is significantly less accurate than results from the LANL system; however, it is still sufficient to allow for some interesting

conclusions. The measured data from the reported experiments used in these analyses are listed in Appendix D.

The NOVA code was capable of distinguishing the light water reactor fuels from the other fuel types for all of the experimental data; however, in some cases the proposed technique had some difficulty determining if the fuel was from a PWR or a BWR. The results for the PWR and BWR fuels are shown in Tables XVI-XIX.

TABLE XVI
Inferred and Declared Burnup Values for the Obrigheim (PWR) Reactor Fuels

			Using xenon only		**Using krypton and xenon**	
Sample	*Enrichment (w/o ^{235}U)*	*Declared Burnup (MWd/MTU)*	*Inferred Burnup (MWd/MTU)*	*Inferred Fuel Type*	*Inferred Burnup (MWd/MTU)*	*Inferred Fuel Type*
86	3.1	28974	26501	PWR	*	*
87	3.1	30917	28683	BWR	33742	PWR
88	3.1	28859	30816	PWR	30594	PWR
89	3.1	29671	25418	BWR	30775	PWR
90	3.1	30181	31390	PWR	31440	PWR
91	3.1	28542	24224	BWR	30492	PWR
92	3.1	27061	27942	PWR	28195	PWR
93	3.1	29857	31359	PWR	31061	PWR
94	3.1	26452	26564	PWR	27382	PWR
95	3.1	28650	28715	PWR	28875	PWR
d1 p1	3.0	19520	19715	PWR	*	*
e3 p2	3.0	29530	29164	PWR	29223	PWR
e3 p4	3.0	30940	30754	PWR	30471	PWR
g7 p1	3.0	17490	13657	BWR	17423	PWR
g7 p3	3.0	31920	33325	PWR	32438	PWR
g7 p5	3.0	28830	28869	PWR	27455	PWR
m14 p1	3.0	15600	15760	PWR	15593	PWR
m14 p4	3.0	27460	28160	PWR	27041	PWR
g14 p31	3.0	37490	38075	PWR	38256	PWR

* No krypton isotopic ratios were reported.

TABLE XVII
Inferred and Declared Burnup Values for the Borssele (PWR) Reactor Fuels

			Using xenon only		Using krypton and xenon	
Sample	*Enrichment (w/o ^{235}U)*	*Declared Burnup (MWd/MTU)*	*Inferred Burnup (MWd/MTU)*	*Inferred Fuel Type*	*Inferred Burnup (MWd/MTU)*	*Inferred Fuel Type*
a8	3.1	31000	31243	PWR	31814	PWR
o6	3.1	31800	32758	PWR	31724	PWR
g15	3.1	31600	32946	PWR	33636	PWR
i15	3.1	31500	32796	PWR	32125	PWR
a10	3.1	31000	33114	PWR	32531	PWR

TABLE XVIII
Inferred and Declared Burnup Values for the Gundremmingen (BWR) Reactor Fuels

			Using xenon only		Using krypton and xenon	
Sample	*Enrichment (w/o ^{235}U)*	*Declared Burnup (MWd/MTU)*	*Inferred Burnup (MWd/MTU)*	*Inferred Fuel Type*	*Inferred Burnup (MWd/MTU)*	*Inferred Fuel Type*
b3	2.53	21240	22063	BWR	22277	BWR
c5	2.53	22970	27562	PWR	23198	BWR
e5	2.53	25190	24623	BWR	24086	BWR
b3	2.53	14390	14710	BWR	18202	PWR
c5	2.53	15840	15547	BWR	19057	PWR
e5	2.53	17490	15790	BWR	20121	PWR

TABLE XIX
Inferred and Declared Burnup Values for the Garigliano (BWR) Reactor Fuels

			Using xenon only		Using krypton and xenon	
Sample	*Enrichment (w/o ^{235}U)*	*Declared Burnup (MWd/MTU)*	*Inferred Burnup (MWd/MTU)*	*Inferred Fuel Type*	*Inferred Burnup (MWd/MTU)*	*Inferred Fuel Type*
a1	1.6	10590	11570	BWR	12187	BWR
a9	1.6	14040	18637	PWR	15297	BWR
b1	1.6	9800	11640	BWR	12480	BWR
j1	1.6	12830	18058	PWR	14959	BWR
j9	1.6	14480	19358	PWR	15883	BWR
a3	2.1	10510	14029	PWR	11556	BWR
b2	2.1	10280	14114	PWR	11452	BWR
b8	2.1	12150	16089	PWR	12960	BWR
c1	2.1	10660	14291	PWR	11692	BWR
c3	2.1	9140	14204	PWR	11387	BWR
d2	2.1	9440	14204	PWR	11398	BWR
d4	2.1	8850	13010	BWR	12963	BWR
e1	2.1	10800	14830	PWR	12173	BWR
e5	2.1	8930	15014	PWR	12268	BWR
g7	2.1	10540	14825	PWR	11886	BWR
h2	2.1	11920	16272	PWR	15739	PWR
h8	2.1	12700	13172	BWR	13971	BWR

The PWR and BWR results were compiled by comparing the number of times the NOVA code correctly predicted the reactor fuel type with xenon isotopes only and using xenon and krypton together. The results of this analysis are shown in Table XX. Table XX lists the percent of correct fuel type predictions for both the PWR and BWR reactors. It is important to note that the technique was significantly more accurate when using the xenon and krypton isotopes together than when using the xenon isotopes alone. This supports the contention that for accurate and consistent safeguards application using both the xenon and krypton isotopes is crucial. Also, the technique was more accurate for the PWRs than the BWRs. Similar results were found when the

accuracy of the burnup determination was analyzed. Comparisons were made between the declared burnup for each sample and the value predicted by the NOVA code. The results of this analysis are shown in Table XXI. Again, the code was more accurate for the PWRs than the BWRs and significantly more accurate when using both the xenon and krypton isotopes compared to when using the xenon isotopes alone.

TABLE XX
Percent of Correct Fuel Type Predictions by the NOVA Code for the LWR Fuels

	PWR	BWR
using xenon only	83.3 %	39.1 %
using xenon and krypton	100.0 %	82.6 %

TABLE XXI
Average Percent Difference Between the Declared Burnup and the Burnup Value Predicted by the NOVA Code for the LWR Fuels

	PWR	BWR
using xenon only	4.93 %	28.33 %
using xenon and krypton	3.25 %	16.70 %

The technique performed consistently better for the PWR fuels than for the BWR fuels. The increased accuracy for PWR reactors is due two characteristics of the samples used in the experiments: sample rod height positions and consistency with the representative model. The monitoring methodology was developed to analyze assembly dissolutions; however, these experiments consisted of samples cut from fuel pins at various rod heights. Thus, for the BWR reactors the moderator density at these different rod heights may have varied by an order of magnitude. For the PWR samples the rod height position of the samples did not significantly impact the moderator density and thus the sample more closely represented an assembly average.

Also, the BWR fuels used in this analysis were markedly different from the representative BWR in the reactor physics database. The representative BWR used 2.75 w/o ^{235}U fuel (Table XIII); however, the fuel samples from the literature had a much lower enrichment. For instance, the Garigliano reactor fuels were 1.6 and 2.1 w/o ^{235}U, and the Gundremmingen fuels were 2.53 w/ ^{235}U. This conclusion is supported by the fact that the technique was more accurate for the Gundremmingen fuels than for the Garigliano fuels. For the Gundremmingen fuels alone, using just the xenon isotopes, the code predicted the correct reactor type 83.3% of the time and estimated the burnup to within 6.65%.

These BWR results were a good test of the technique since the fuels are clearly different from the average BWR. This implies that even for fuels with lower enrichment than the representative average in the database, or perhaps incomplete fuel

chopping, the technique still results in a reasonably accurate prediction of the burnup and fuel type (especially if both the xenon and krypton isotopic ratios are used).

Conversely, the PWR fuels analyzed were reasonably similar to the representative average. Both of the PWRs had enrichments of 3.0 or 3.1 w/o ^{235}U, whereas the representative average in the database had an enrichment of 2.85 w/o ^{235}U. This resulted in excellent performance in both the burnup and fuel type prediction for the Inverse Problem.

Some of the literature measurements (specifically the Borssele fuel and the 3.0 w/o ^{235}U Obrigheim fuel) had reported values for the $^{85}Kr/^{86}Kr$ isotopic ratios. This provided an opportunity to analyze the NOVA code's ability to predict fuel age. The fuel age is determined using the technique described in Section V above. The Borssele fuel was 2.85 years old at the time of the experiment and all fuel samples were analyzed at the same time. The NOVA code predicted an average fuel age (averaged over all samples) of 2.88 ± 0.16 years for the Borssele fuel. The Obrigheim fuel was 3.92 years old at the time of the experiment and all fuel samples were analyzed at the same time. The NOVA code predicted an average fuel age (averaged over all samples) of 3.84 ± 0.32 years for the Obrigheim fuel. These results demonstrate that the NOVA code and methodology are sufficiently accurate for determining fuel age even from less than exceptional measurements. The accuracy of these predictions was more than sufficient for establishing the cycle after which the fuel was discharged.

These results suggest that the proposed technique, including the reactor physics databases, is valid for use with LWR fuels for the Inverse Problem and can reasonably

distinguish the minute difference between PWR and BWR fuels. Also, it is expected that the technique developed will result in even more accurate predictions when used in the Forward Problem. The measurements reported in the literature were not of the same accuracy as that acquired using the LANL spectrometer; thus, it is expected that if similar samples were analyzed using the LANL spectrometer for the LWR fuels better predictions would result.

C. Operational History Determinations

Thus far the proposed technique has been developed for the prediction of fuel type, burnup, and fuel age. It was noted in Section II that the $^{136}Xe/^{134}Xe$ isotopic ratios contain information regarding the operational history of the reactor. This operational history includes information such as total reactor power level versus time and assembly position (or power peaking factor) versus time. Since the $^{136}Xe/^{134}Xe$ isotopic ratio is affected by several operational parameters, it was deemed unsuitable for use in the Inverse Problem; however, it is feasible that this ratio could be used to aid in confirming declared power histories through a Forward Problem analysis. In this role, an investigator could model an existing reactor using a declared power profile (such as specific power for the fuel versus time) and compare the measured $^{136}Xe/^{134}Xe$ isotopic ratio to that suggested by the calculation. Thus, the probability of measuring a certain value of the ratio could be determined using the following approach:

$$p(R_6^m \mid M,B,E) = \frac{1}{\sigma_6^c\sqrt{2\pi}}\exp\left[-\frac{(R_6^m - R_6^c)^2}{2(\sigma_6^c)^2}\right]. \qquad (56)$$

where R_6^m is the measured $^{136}Xe/^{134}Xe$ isotopic ratio, R_6^c is the calculated $^{136}Xe/^{134}Xe$ for model M at burnup B, and σ_6^c is the standard deviation of R_6^c. The standard deviation of the calculated isotopic ratios would be determined using a series of benchmark experiments and from an estimation of the accuracy of the declared information (i.e., power profile).

To examine the potential use of this technique, simulations were performed to estimate the sensitivity of the $^{136}Xe/^{134}Xe$ isotopic ratio to simple changes in power level. Reactor physics calculations were performed to determine the isotopic composition of spent fuel from a Calder Hall reactor when the fuel is exposed for two different power histories (Figs. 38-39). Both fuels achieved the same final burnup (2000 MWd/MTU). The HELIOS lattice physics code was used for all of the calculations. The two power histories used were a Constant Power Event and an Up Power Event. For the Constant Power Event, the fuel was exposed at a constant power of 1.76 W/g (100% of full power) from 0 to 2000 MWd/MTU. For the Up Power Event, the fuel was exposed at a constant power of 0.88 W/g (50% of full power) from 0 to 1000 MWd/MTU. Then the power was increased, and the fuel was exposed at a constant power of 1.76 W/g (100% of full power) from 1000 to 2000 MWd/MTU.

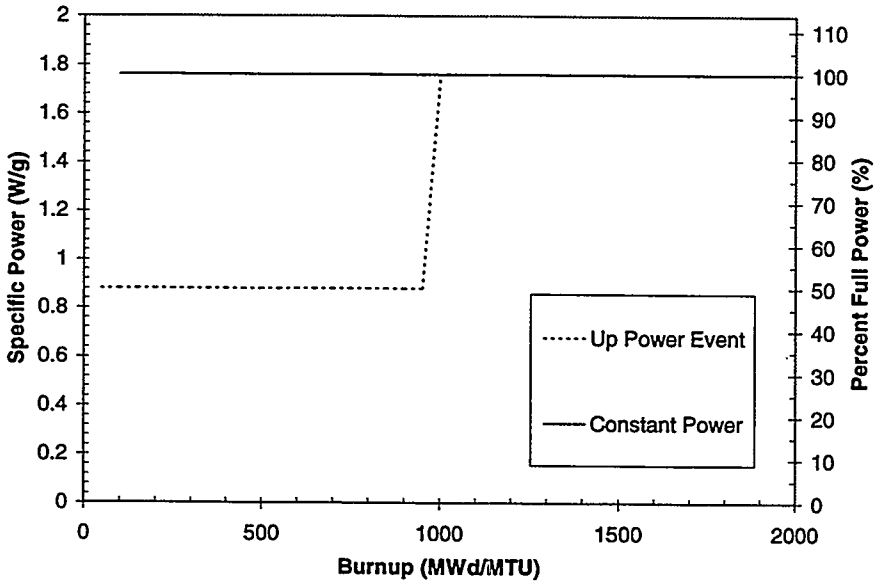

Fig. 38. Two power histories for use with the Calder Hall reactor illustrated through power versus burnup plots.

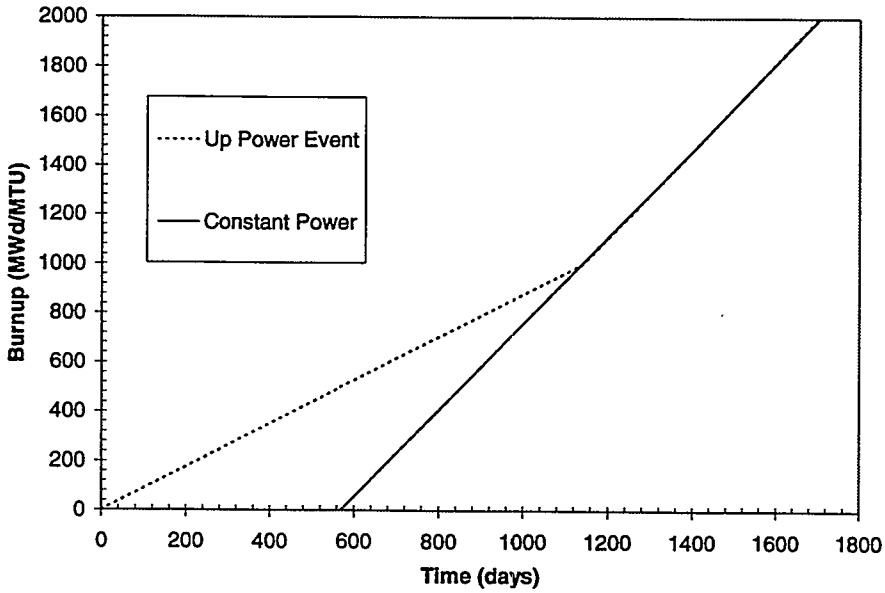

Fig. 39. Two power histories for use with the Calder Hall reactor illustrated through burnup versus time plots.

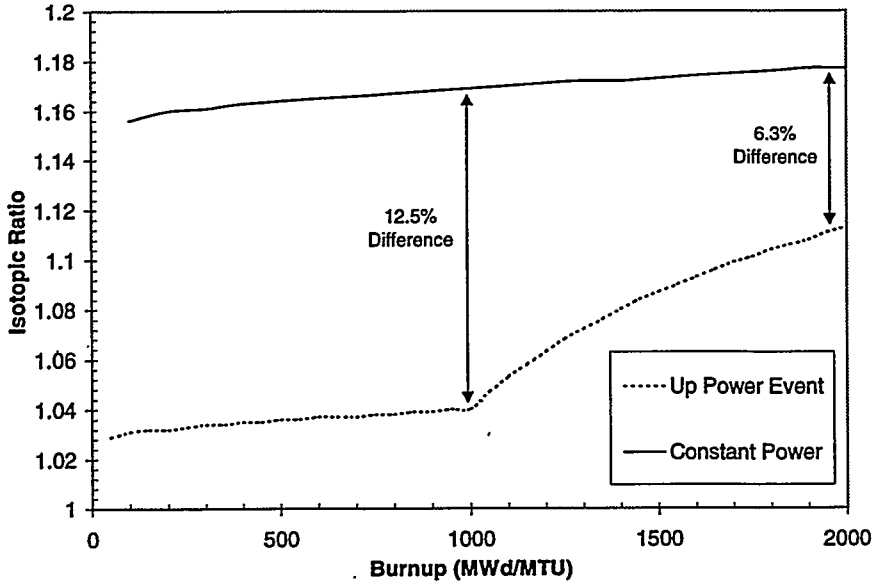

Fig. 40. $^{136}Xe/^{134}Xe$ isotopic ratio versus burnup for Calder Hall reactor using two power histories.

Significant changes were noted in the calculated $^{136}Xe/^{134}Xe$ isotopic ratio in the spent fuel at 2000 MWd/MTU for the two power histories (Fig. 40). The Up Power Event history fuel has a $^{136}Xe/^{134}Xe$ isotopic ratio approximately 6.3% lower than that of the Constant Power history fuel at the final burnup value of 2000 MWd/MTU. Distinguishing this difference should be well within the measurement capabilities of even a portable mass spectrometry system. The calculated $^{136}Xe/^{134}Xe$ isotopic ratios are estimated to be accurate to within a single standard deviation of ±1-2%. Full development of this methodology would be performed on a case-by-case basis, especially since the HELIOS lattice physics code is not commercially available in a PC

version and would therefore be difficult to transport to a particular site. The brief description here is only meant to demonstrate the potential use of this isotopic ratio, and it is not included in the NOVA code.

VII. CONCLUSIONS AND RECOMMENDATIONS

An accurate and noninvasive environmental monitoring technique, which integrated existing technologies to strengthen safeguards at reprocessing facilities, was developed. This technique involves the measurement of isotopic ratios of stable noble fission gases from on-stack emissions during reprocessing of spent fuel using high-precision mass spectrometry. These results were then 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). The calculated database was well benchmarked for many reactor types.

The complete system (mass spectrometry, reactor modeling, and data analysis) was validated using on-stack measurements during the reprocessing of low-burnup target fuel from a U.S. production reactor and gas samples from the processing of high-burnup fast breeder reactor driver fuel. These measurements led to an inferred burnup that matched the declared burnup with sufficient accuracy and consistency for most safeguards applications. The NOVA code was also 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 xenon and krypton isotopic ratios). In all, NOVA is capable of determining all of the following characteristics for any reprocessed fuel:

1. Distinguish low-burnup from high-burnup levels,
2. Determine the spent fuel burnup (within ~ 4%),
3. Determine the reactor type that produced the fuel,
4. Determine the fuel age (with ~4%),
5. Determine the $^{240}Pu/^{239}Pu$ isotopic ratio for the spent fuel,
6. Determine the plutonium content of the spent fuel,
7. 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 plutonium content and the determination of the concentrations of other fission products. A methodology was also developed for confirming declared power histories, but this was not incorporated into the NOVA code.

The technique generated here has been validated for use in many systems, but the monitoring technique is not foolproof. Proper implementation in a safeguards regime would still require inspection of the reprocessing procedures and gas system piping. This would aid in detering any potential diversion or mixing of the exhaust gas to conceal proliferation activities. This technique is intended for use in a strengthened safeguards setting and may also be helpful when implemented in a voluntary manner by

any facility or nation wishing to demonstrate that no weapons-useable material is being produced.

Continued validation is necessary to demonstrate the robustness of this system for production, power, and research reactor fuels (esp., for graphite-moderated systems). The existing reactor physics database has not been tested for graphite-moderated fuels and any results for these types of reactors should be suspect prior to proper validation. Also, continued validation for MOX fuels and CANDU fuels should be performed; however, this is likely to require significant collaboration with foreign organizations. The existing MOX models may be overly simplified. After further validation, it may be decided to include multiple MOX fuel designs (esp. to distinguish between weapons-grade and reactor grade MOX fuels or different mixtures of MOX and LEU fuels).

Some suggestions were made on the effective application of this methodology. Perhaps the most promising usage 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 the plutonium in the fuel. Other strategies for the application of this technique will be left for exploration by the user.

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APPENDIX A

Reactor Physics Code Benchmark Results

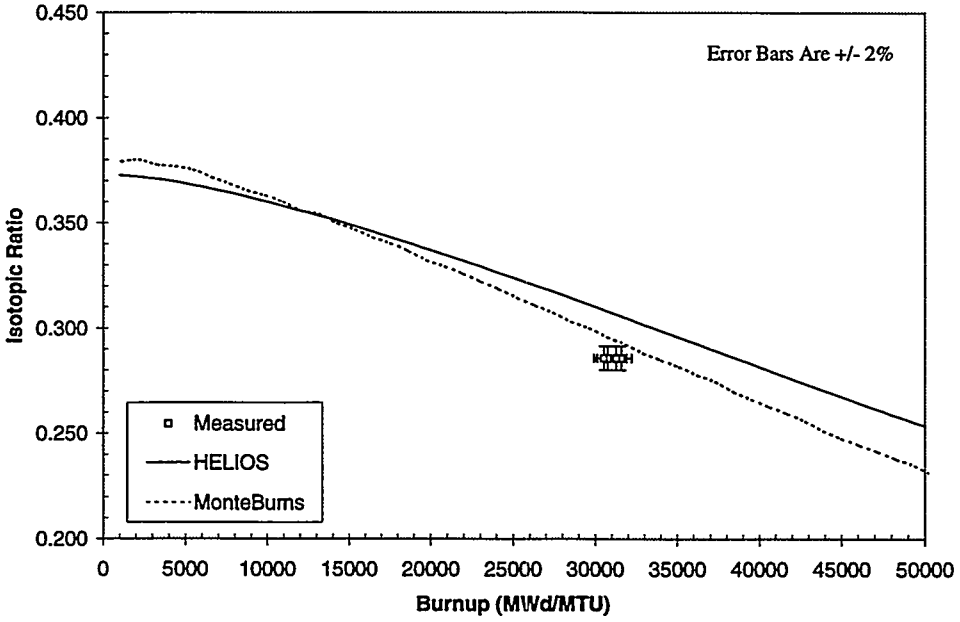

Fig. A.1. $^{131}Xe/^{134}Xe$ isotopic ratio versus burnup for the Turkey Point Reactor (2.56 w/o ^{235}U fuel).

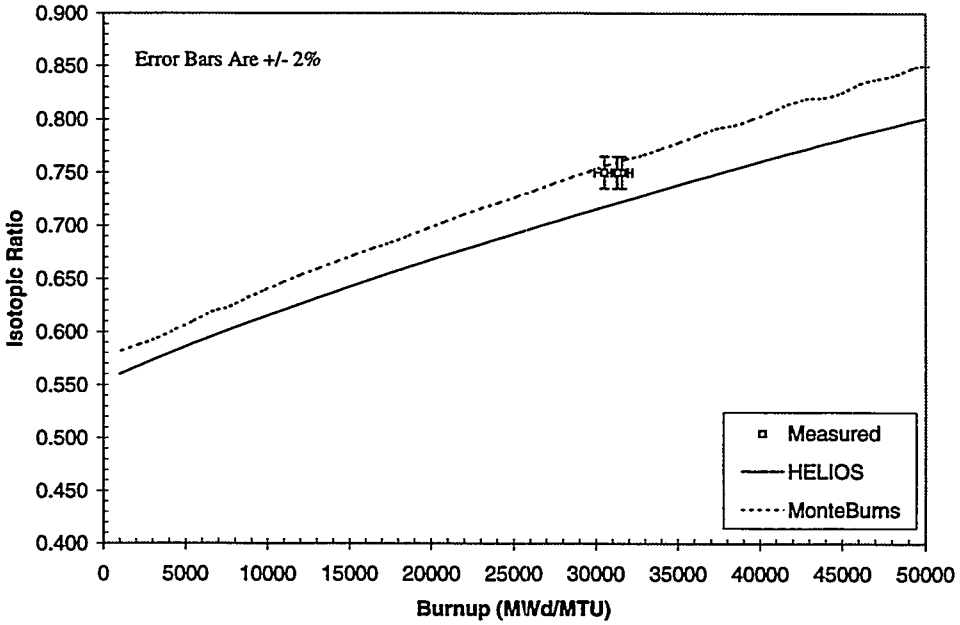

Fig. A.2. $^{132}Xe/^{134}Xe$ isotopic ratio versus burnup for the Turkey Point Reactor (2.56 w/o ^{235}U fuel).

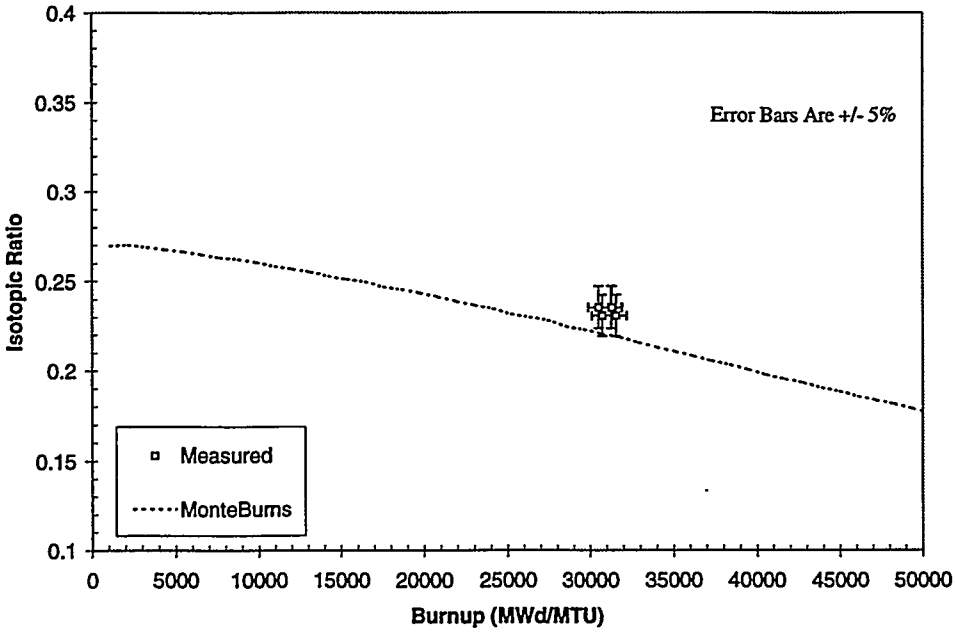

Fig. A.3. $^{83}Kr/^{86}Kr$ isotopic ratio versus burnup for the Turkey Point Reactor (2.56 w/o ^{235}U fuel).

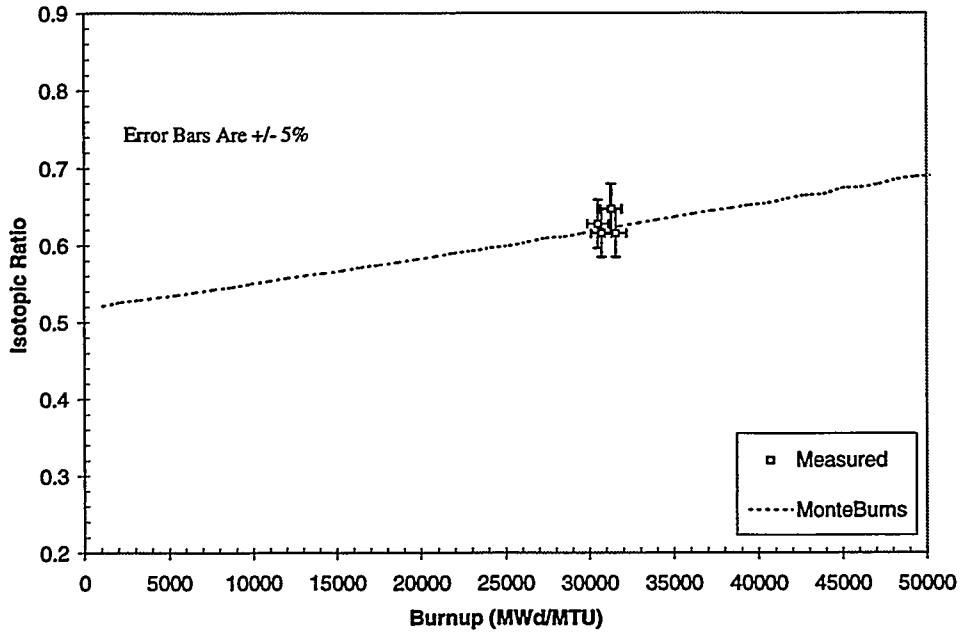

Fig. A.4. $^{84}Kr/^{86}Kr$ isotopic ratio versus burnup for the Turkey Point Reactor (2.56 w/o ^{235}U fuel).

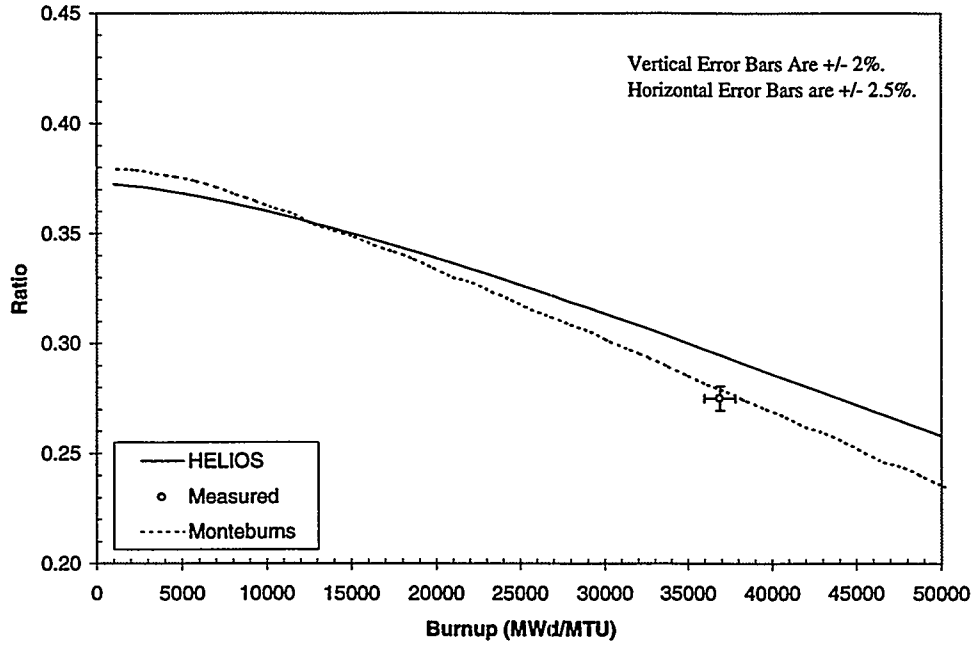

Fig. A.5. $^{131}Xe/^{134}Xe$ isotopic ratio versus burnup for the Obrigheim Reactor (2.83 w/o ^{235}U fuel).

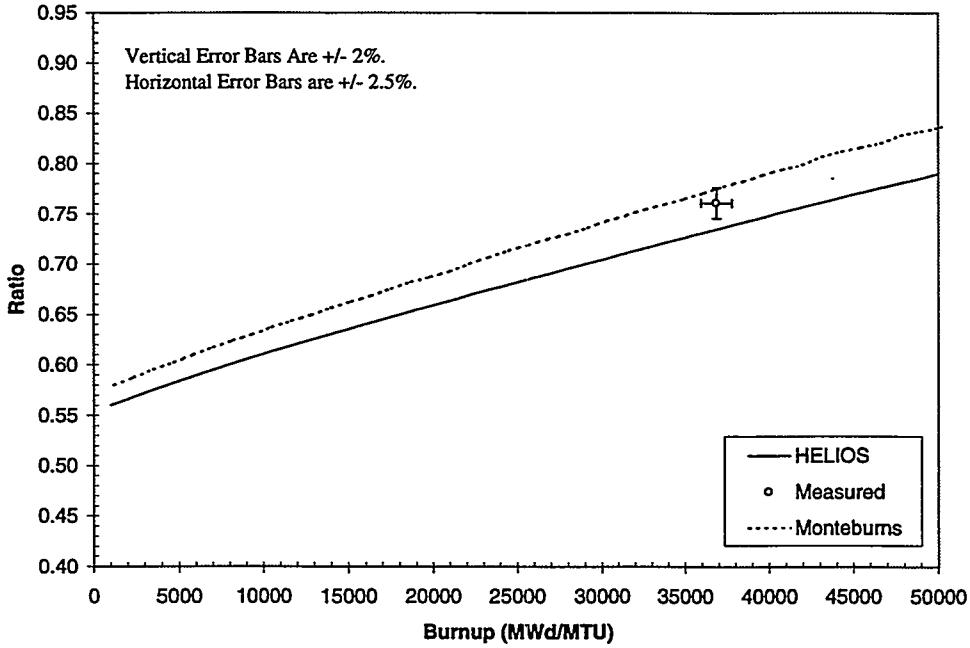

Fig. A.6. $^{132}Xe/^{134}Xe$ isotopic ratio versus burnup for the Obrigheim Reactor (2.83 w/o ^{235}U fuel).

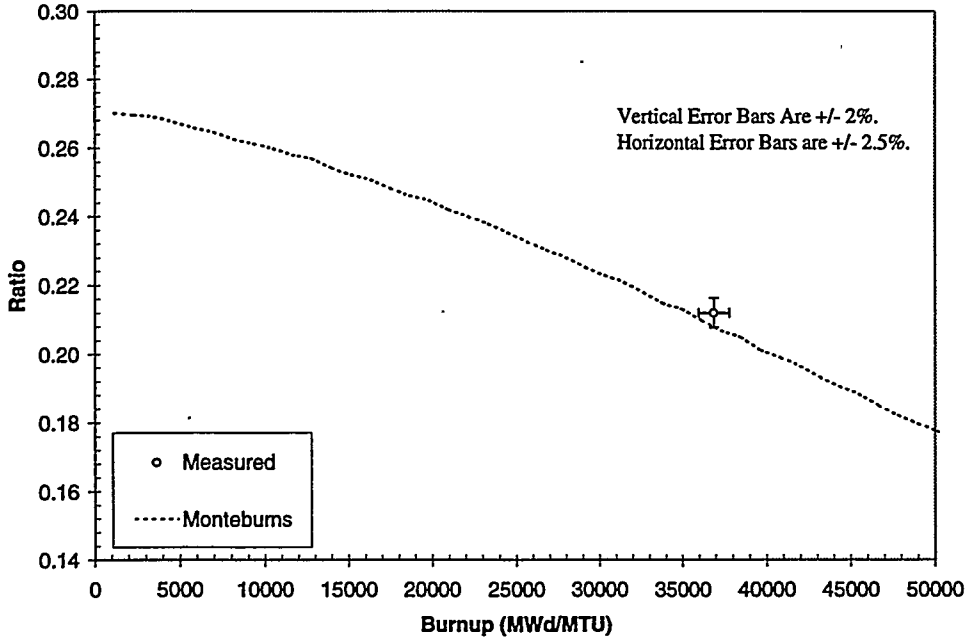

Fig. A.7. $^{83}Kr/^{86}Kr$ isotopic ratio versus burnup for the Obrigheim Reactor (2.83 w/o ^{235}U fuel).

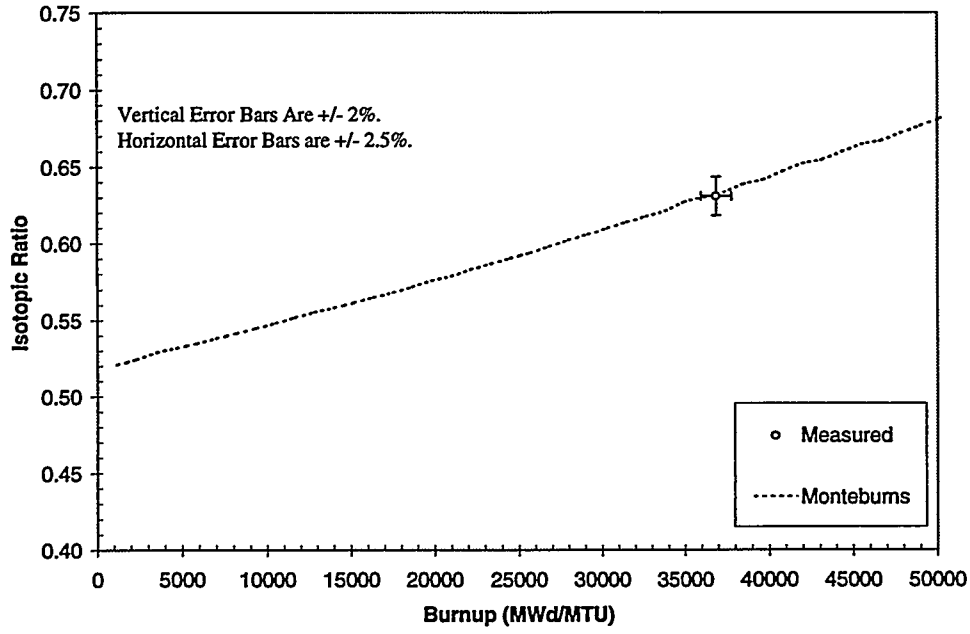

Fig. A.8. $^{84}Kr/^{86}Kr$ isotopic ratio versus burnup for the Obrigheim Reactor (2.83 w/o ^{235}U fuel).

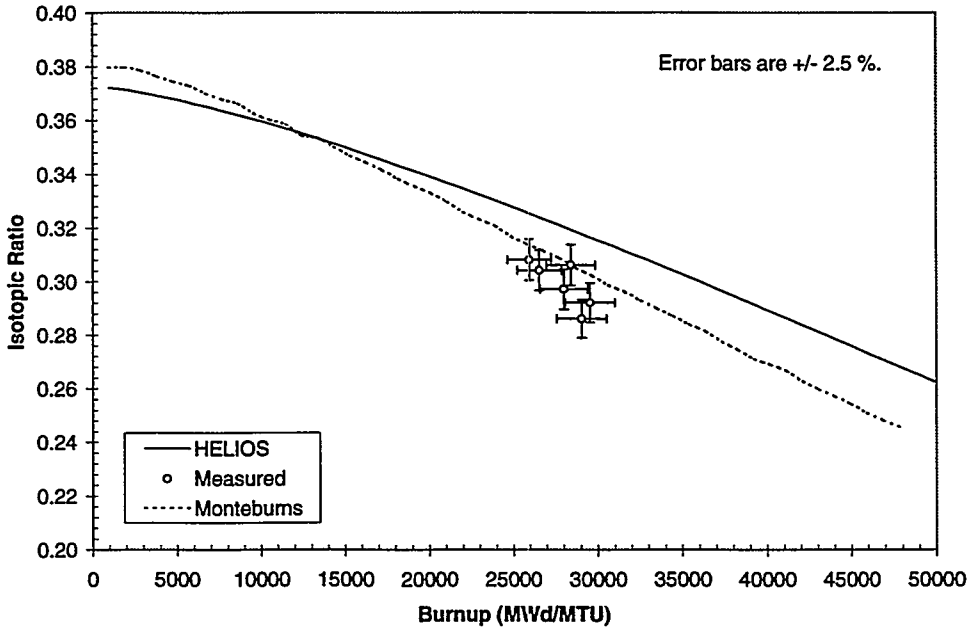

Fig. A.9. $^{131}Xe/^{134}Xe$ isotopic ratio versus burnup for the Obrigheim Reactor (3.13 w/o ^{235}U fuel).

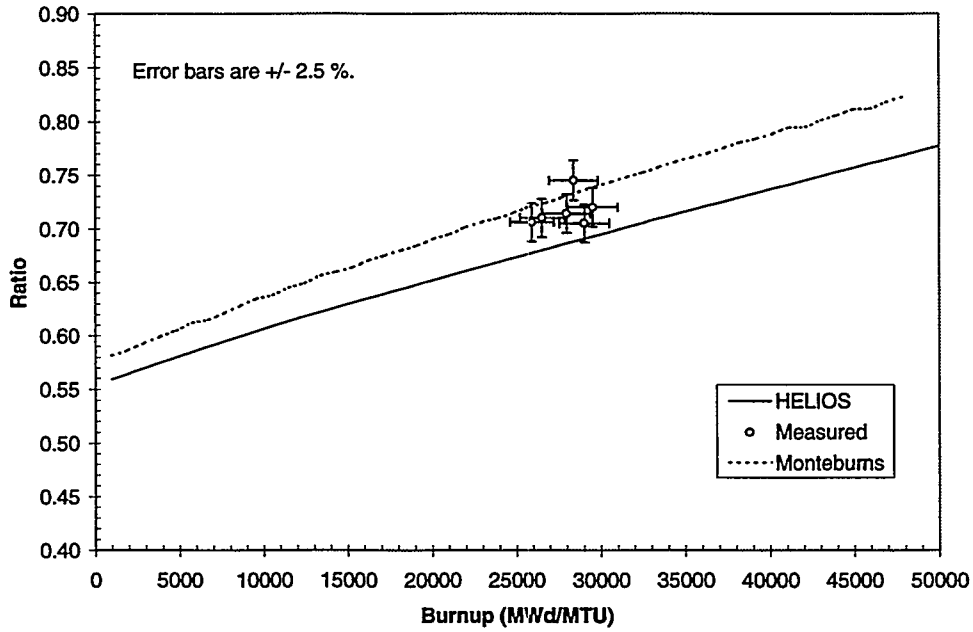

Fig. A.10. $^{132}Xe/^{134}Xe$ isotopic ratio versus burnup for the Obrigheim Reactor (3.13 w/o ^{235}U fuel).

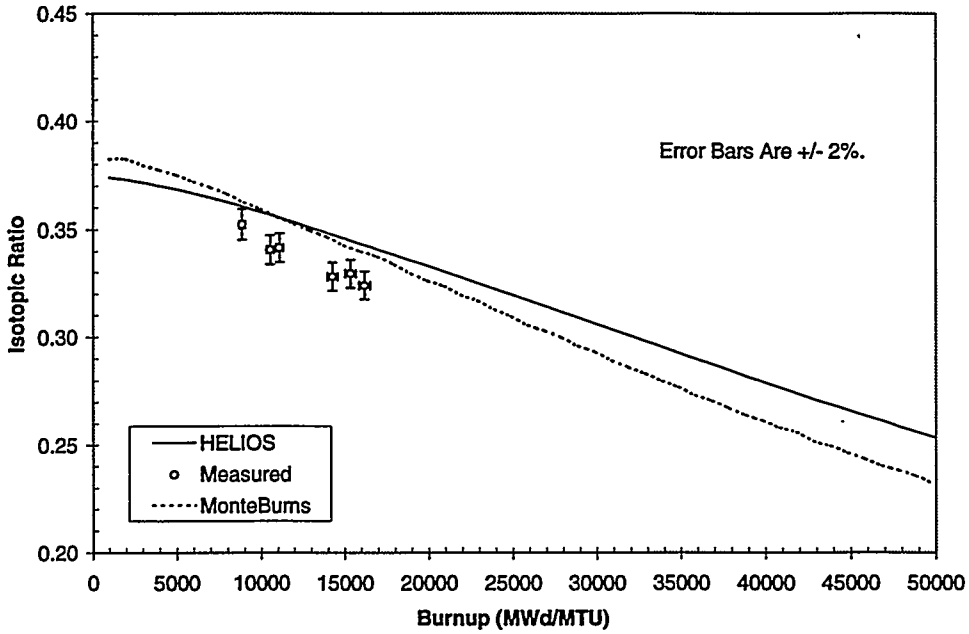

Fig. A.11. $^{131}Xe/^{134}Xe$ isotopic ratio versus burnup for the Trino Vercellese Reactor (2.72 w/o ^{235}U fuel).

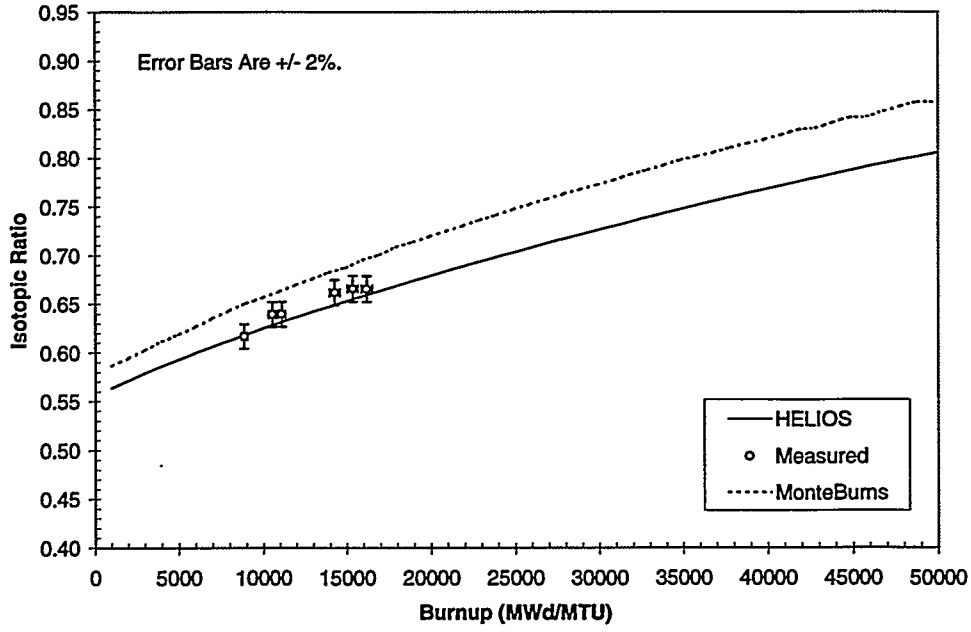

Fig. A.12. $^{132}Xe/^{134}Xe$ isotopic ratio versus burnup for the Trino Vercellese Reactor (2.72 w/o ^{235}U fuel).

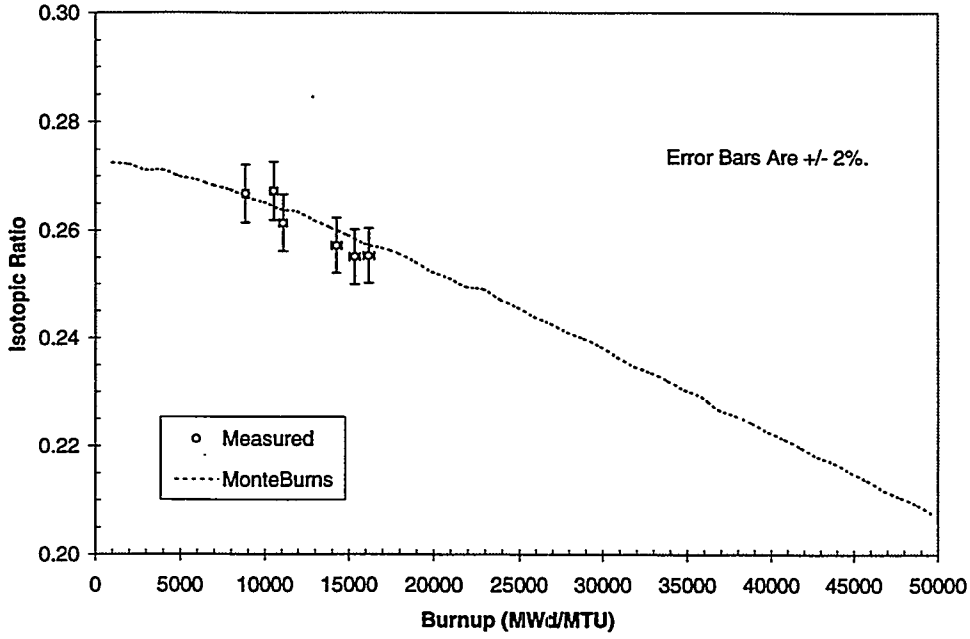

Fig. A.13. $^{83}Kr/^{86}Kr$ isotopic ratio versus burnup for the Trino Vercellese Reactor (2.72 w/o ^{235}U fuel).

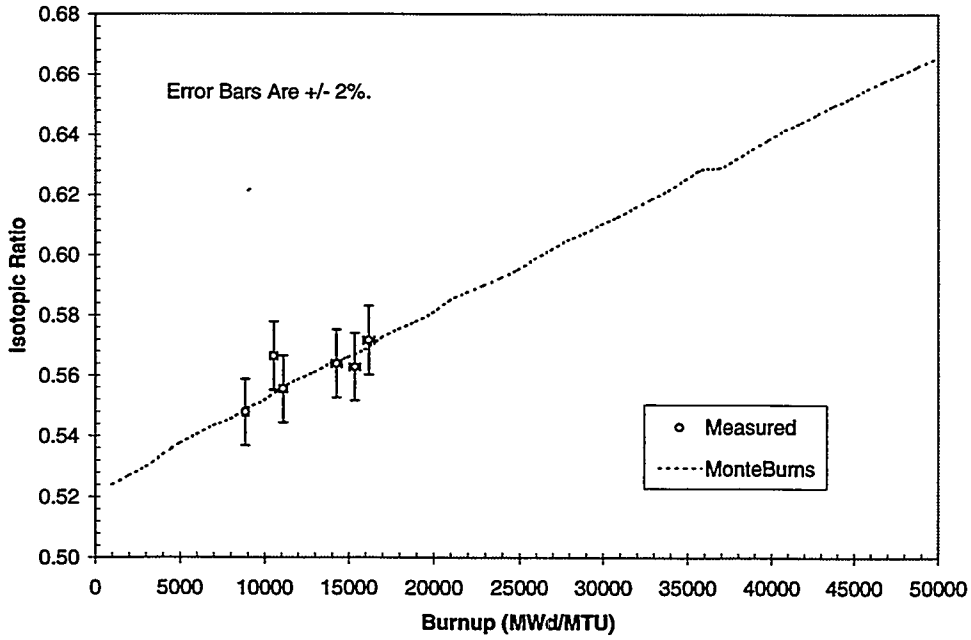

Fig. A.14. $^{84}Kr/^{86}Kr$ isotopic ratio versus burnup for the Trino Vercellese Reactor (2.72 w/o ^{235}U fuel).

APPENDIX B

Monteburns Input Decks for Various Reactor Fuel Types in the Databases

PWR-LEU Decks

```
Average PWR-LEU Pin Cell with Correct FM Ratio for MCNP
1     1 -10.000  -1     -16  17              imp:n=1 $Fuel Zone 1
2     2 -10.000  -2    1 -16  17              imp:n=1 $Fuel Zone 2
3     3 -10.000  -3    2 -16  17              imp:n=1 $Fuel Zone 3
4     4 -10.000  -4    3 -16  17              imp:n=1 $Fuel Zone 4
5     5 -10.000  -5    4 -16  17              imp:n=1 $Fuel Zone 5
6     6 -10.000  -6    5 -16  17              imp:n=1 $Fuel Zone 6
7     7 -10.000  -7    6 -16  17              imp:n=1 $Fuel Zone 7
8     8 -10.000  -8    7 -16  17              imp:n=1 $Fuel Zone 8
9     9 -10.000  -9    8 -16  17              imp:n=1 $Fuel Zone 9
10   10 -6.5600 -10    9 -16  17              imp:n=1 $Clad
11   11 -0.7027  10 -16  17 -12  13 -14  15 imp:n=1 $Coolant
12    0          16                          imp:n=0 $Universe Above Pin
13    0         -17                          imp:n=0 $Universe Below Pin
14    0         -16  17  12                  imp:n=0 $Universe Top Pin
15    0         -16  17 -13                  imp:n=0 $Universe Bot Pin
16    0         -16  17 -12  13  14          imp:n=0 $Universe Right Pin
17    0         -16  17 -12  13 -15          imp:n=0 $Universe Left Pin

1     cz 0.199
2     cz 0.313
3     cz 0.379
4     cz 0.417
5     cz 0.439
6     cz 0.452
7     cz 0.459
8     cz 0.464
9     cz 0.466                                  $Fuel Outer Radius
10    cz 0.536                                  $Clad Outer Radius
*12   px 0.7492                                 $P/2
*13   px -0.7492
*14   py 0.7492
*15   py -0.7492
*16   pz 209.402                                $H/2
*17   pz -209.402

mode:n
kcode 8000 1.0 50 450
ksrc 0.000 0.0 0.0
m1    8016.60c  4.4614E-02
      92234.60c 5.7530E-06
      92235.54c 6.4364E-04
      92236.60c 2.9482E-06
      92238.54c 2.1655E-02                    $UO2
m2    8016.60c  4.4614E-02
      92234.60c 5.7530E-06
      92235.54c 6.4364E-04
      92236.60c 2.9482E-06
      92238.54c 2.1655E-02                    $UO2
m3    8016.60c  4.4614E-02
      92234.60c 5.7530E-06
      92235.54c 6.4364E-04
      92236.60c 2.9482E-06
      92238.54c 2.1655E-02                    $UO2
m4    8016.60c  4.4614E-02
      92234.60c 5.7530E-06
      92235.54c 6.4364E-04
      92236.60c 2.9482E-06
      92238.54c 2.1655E-02                    $UO2
m5    8016.60c  4.4614E-02
      92234.60c 5.7530E-06
      92235.54c 6.4364E-04
      92236.60c 2.9482E-06
      92238.54c 2.1655E-02                    $UO2
```

```
m6    8016.60c  4.4614E-02
      92234.60c 5.7530E-06
      92235.54c 6.4364E-04
      92236.60c 2.9482E-06
      92238.54c 2.1655E-02                    $UO2
m7    8016.60c  4.4614E-02
      92234.60c 5.7530E-06
      92235.54c 6.4364E-04
      92236.60c 2.9482E-06
      92238.54c 2.1655E-02                    $UO2
m8    8016.60c  4.4614E-02
      92234.60c 5.7530E-06
      92235.54c 6.4364E-04
      92236.60c 2.9482E-06
      92238.54c 2.1655E-02                    $UO2
m9    8016.60c  4.4614E-02
      92234.60c 5.7530E-06
      92235.54c 6.4364E-04
      92236.60c 2.9482E-06
      92238.54c 2.1655E-02                    $UO2
m10   24000.50c 7.5976E-05
      26000.55c 1.5562E-04
      40000.60c 4.2605E-02
      50000.35c 4.3269E-04                    $Zircaloy-4
m11   1001.60c  4.6978E-02
      8016.60c  2.3489E-02                    $H2O
mt11  lwtr.04t
tmp   7.76E-08
      7.76E-08
      7.76E-08
      7.76E-08
      7.76E-08
      7.76E-08
      7.76E-08
      7.76E-08
      7.76E-08
      5.26E-08
      4.83E-08
      0.00E-00
      0.00E-00
      0.00E-00
      0.00E-00
      0.00E-00
      0.00E-00                                $Temperatures
xs1   35082.00c
      81.213094
      acebr82
      ~/Codes/MCNP/xsections
      1
      1
      1911
      0
      0
      2.585E-08
xs2   35083.00c
      82.202898
      acebr83
      ~/Codes/MCNP/xsections
      1
      1
      1911
      0
      0
      2.585E-08
xs3   36085.00c
      84.183100
      acekr85
      ~/Codes/MCNP/xsections
```

```
     1
     1
     2556
     0
     0
     2.585E-08
xs4  52131.00c
     129.781566
     acete131
     ~/Codes/MCNP/xsections
     1
     1
     1911
     0
     0
     2.585E-08
xs5  52132.00c
     130.775000
     acete132
     ~/Codes/MCNP/xsections
     1
     1
     2779
     0
     0
     2.585E-08
xs6  53130.00c
     128.791000
     acei130
     ~/Codes/MCNP/xsections
     1
     1
     2657
     0
     0
     2.585E-08
xs7  53131.00c
     129.782000
     acei131
     ~/Codes/MCNP/xsections
     1
     1
     2943
     0
     0
     2.585E-08
xs8  53132.00c
     130.774832
     acei132
     ~/Codes/MCNP/xsections
     1
     1
     1911
     0
     0
     2.585E-08
xs9  54130.00c
     128.788000
     acexe130
     ~/Codes/MCNP/xsections
     1
     1
     21096
     0
     0
     2.585E-08
xs10 54132.00c
     130.771000
```

```
     acexe132
     ~/Codes/MCNP/xsections
     1
     1
     9937
     0
     0
     2.585E-08
xs11 54136.00c
     134.740000
     acexe136
     ~/Codes/MCNP/xsections
     1
     1
     4429
     0
     0
     2.585E-08
print 40
```

```
Average PWR-LEU Monteburns Input File
9
1
2
3
4
5
6
7
8
9
0.0
0.0
0.0
0.0
0.0
0.0
0.0
0.0
0.0
0.100000
-200.0
2000
35
10
1
0
22
/nh/whirlpool/u/wscharl/Codes/Origen/Libs
0.005
0
28
28
28
28
28
28
28
28
28
28
28
28
28
28
28
28
28
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
```

```
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53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
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54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
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53130.00c
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53132.00c
54130.00c
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54134.35c
54136.00c
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94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
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36082.50c
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53132.00c
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54136.00c
92234.60c
92235.54c
```

92236.60c
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94238.60c
94239.60c
94240.60c
94241.60c
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35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
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52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
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92235.54c
92236.60c
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35083.00c
36082.50c
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36085.00c
36086.50c
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94240.60c
94241.60c
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35083.00c
36082.50c

```
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36084.50c
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52132.00c
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53132.00c
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94239.60c
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36082.50c
36083.50c
36084.50c
36085.00c
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35083.00c
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36083.50c
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53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
```

```
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92238.54c
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93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
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53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
```

BWR-LEU Decks

```
Average BWR-LEU Pin Cell with Correct FM Ratio
1     1 -10.000  -1      -16  17                imp:n=1 $Fuel Zone 1
2     2 -10.000  -2    1 -16  17                imp:n=1 $Fuel Zone 2
3     3 -10.000  -3    2 -16  17                imp:n=1 $Fuel Zone 3
4     4 -10.000  -4    3 -16  17                imp:n=1 $Fuel Zone 4
5     5 -10.000  -5    4 -16  17                imp:n=1 $Fuel Zone 5
6     6 -10.000  -6    5 -16  17                imp:n=1 $Fuel Zone 6
7     7 -10.000  -7    6 -16  17                imp:n=1 $Fuel Zone 7
8     8 -10.000  -8    7 -16  17                imp:n=1 $Fuel Zone 8
9     9 -10.000  -9    8 -16  17                imp:n=1 $Fuel Zone 9
10   10 -6.5600 -10    9 -16  17                imp:n=1 $Clad
11   11 -0.4500  10 -16  17 -12  13 -14  15     imp:n=1 $Coolant
12    0           16                            imp:n=0 $Universe Above Pin
13    0          -17                            imp:n=0 $Universe Below Pin
14    0          -16  17  12                    imp:n=0 $Universe Top Pin
15    0          -16  17 -13                    imp:n=0 $Universe Bottom Pin
16    0          -16  17 -12  13  14            imp:n=0 $Universe Right Pin
17    0          -16  17 -12  13 -15            imp:n=0 $Universe Left Pin

1     cz 0.264
2     cz 0.417
3     cz 0.505
4     cz 0.555
5     cz 0.585
6     cz 0.601
7     cz 0.611
8     cz 0.617
9     cz 0.620                                  $Fuel Outer Radius
10    cz 0.715                                  $Clad Outer Radius
*12   px 1.0150                                 $P/2
*13   px -1.0150
*14   py 1.0150
*15   py -1.0150
*16   pz 165.6135                               $H/2
*17   pz -165.6135

mode:n
kcode 8000 1.0 50 450
ksrc 0.000 0.0 0.0
m1    8016.60c  4.4614E-02
      92234.60c 6.0557E-06
      92235.54c 6.7752E-04
      92236.60c 3.1033E-06
      92238.54c 2.1621E-02                      $UO2
m2    8016.60c  4.4614E-02
      92234.60c 6.0557E-06
      92235.54c 6.7752E-04
      92236.60c 3.1033E-06
      92238.54c 2.1621E-02                      $UO2
m3    8016.60c  4.4614E-02
      92234.60c 6.0557E-06
      92235.54c 6.7752E-04
      92236.60c 3.1033E-06
      92238.54c 2.1621E-02                      $UO2
m4    8016.60c  4.4614E-02
      92234.60c 6.0557E-06
      92235.54c 6.7752E-04
      92236.60c 3.1033E-06
      92238.54c 2.1621E-02                      $UO2
m5    8016.60c  4.4614E-02
      92234.60c 6.0557E-06
      92235.54c 6.7752E-04
      92236.60c 3.1033E-06
      92238.54c 2.1621E-02                      $UO2
```

```
m6    8016.60c  4.4614E-02
      92234.60c 6.0557E-06
      92235.54c 6.7752E-04
      92236.60c 3.1033E-06
      92238.54c 2.1621E-02                    $UO2
m7    8016.60c  4.4614E-02
      92234.60c 6.0557E-06
      92235.54c 6.7752E-04
      92236.60c 3.1033E-06
      92238.54c 2.1621E-02                    $UO2
m8    8016.60c  4.4614E-02
      92234.60c 6.0557E-06
      92235.54c 6.7752E-04
      92236.60c 3.1033E-06
      92238.54c 2.1621E-02                    $UO2
m9    8016.60c  4.4614E-02
      92234.60c 6.0557E-06
      92235.54c 6.7752E-04
      92236.60c 3.1033E-06
      92238.54c 2.1621E-02                    $UO2
m10   24000.50c 7.5976E-05
      26000.55c 1.5562E-04
      40000.60c 4.2605E-02
      50000.35c 4.3269E-04                    $Zircaloy-4
m11   1001.60c  3.0084E-02
      8016.60c  1.5042E-02                    $H2O
mt11  lwtr.04t
tmp   7.76E-08
      7.76E-08
      7.76E-08
      7.76E-08
      7.76E-08
      7.76E-08
      7.76E-08
      7.76E-08
      7.76E-08
      5.26E-08
      4.83E-08
      0.00E-00
      0.00E-00
      0.00E-00
      0.00E-00
      0.00E-00
      0.00E-00                                $Temperatures
xs1   35082.00c
      81.213094
      acebr82
      ~/Codes/MCNP/xsections
      1
      1
      1911
      0
      0
      2.585E-08
xs2   35083.00c
      82.202898
      acebr83
      ~/Codes/MCNP/xsections
      1
      1
      1911
      0
      0
      2.585E-08
xs3   36085.00c
      84.183100
      acekr85
      ~/Codes/MCNP/xsections
```

```
      1
      1
      2556
      0
      0
      2.585E-08
xs4   52131.00c
      129.781566
      acete131
      ~/Codes/MCNP/xsections
      1
      1
      1911
      0
      0
      2.585E-08
xs5   52132.00c
      130.775000
      acete132
      ~/Codes/MCNP/xsections
      1
      1
      2779
      0
      0
      2.585E-08
xs6   53130.00c
      128.791000
      acei130
      ~/Codes/MCNP/xsections
      1
      1
      2657
      0
      0
      2.585E-08
xs7   53131.00c
      129.782000
      acei131
      ~/Codes/MCNP/xsections
      1
      1
      2943
      0
      0
      2.585E-08
xs8   53132.00c
      130.774832
      acei132
      ~/Codes/MCNP/xsections
      1
      1
      1911
      0
      0
      2.585E-08
xs9   54130.00c
      128.788000
      acexe130
      ~/Codes/MCNP/xsections
      1
      1
      21096
      0
      0
      2.585E-08
xs10  54132.00c
      130.771000
```

```
      acexe132
      ~/Codes/MCNP/xsections
      1
      1
      9937
      0
      0
      2.585E-08
xs11 54136.00c
      134.740000
      acexe136
      ~/Codes/MCNP/xsections
      1
      1
      4429
      0
      0
      2.585E-08
print 40
```

```
Average BWR-LEU Monteburns Input File
9
1
2
3
4
5
6
7
8
9
0.0
0.0
0.0
0.0
0.0
0.0
0.0
0.0
0.0
0.100000
-200.0
2000
25
10
1
10
30
/nh/whirlpool/u/wscharl/Codes/Origen/Libs
0.005
0
28
28
28
28
28
28
28
28
28
28
28
28
28
28
28
28
28
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
```

```
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
```

```
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
```

```
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
```

```
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
```

CANDU-Nat.U Decks

```
Average CANDU-Nat.U Pin Cell with Correct FM Ratio for MCNP
1     1 -10.000      -1     -16  17              imp:n=1 $Fuel Zone 1
2     2 -10.000      -2    1 -16  17              imp:n=1 $Fuel Zone 2
3     3 -10.000      -3    2 -16  17              imp:n=1 $Fuel Zone 3
4     4 -10.000      -4    3 -16  17              imp:n=1 $Fuel Zone 4
5     5 -10.000      -5    4 -16  17              imp:n=1 $Fuel Zone 5
6     6 -10.000      -6    5 -16  17              imp:n=1 $Fuel Zone 6
7     7 -10.000      -7    6 -16  17              imp:n=1 $Fuel Zone 7
8     8 -10.000      -8    7 -16  17              imp:n=1 $Fuel Zone 8
9     9 -10.000      -9    8 -16  17              imp:n=1 $Fuel Zone 9
10   10 -6.5600     -10    9 -16  17              imp:n=1 $Clad
11   11 9.9756E-02   10 -16  17 -12  13 -14  15 imp:n=1 $Coolant
12    0              16                          imp:n=0 $Universe Above Pin
13    0             -17                          imp:n=0 $Universe Below Pin
14    0             -16  17  12                  imp:n=0 $Universe Top Pin
15    0             -16  17 -13                  imp:n=0 $Universe Bottom Pin
16    0             -16  17 -12  13  14          imp:n=0 $Universe Right Pin
17    0             -16  17 -12  13 -15          imp:n=0 $Universe Left Pin

1     cz 0.199
2     cz 0.313
3     cz 0.379
4     cz 0.417
5     cz 0.439
6     cz 0.452
7     cz 0.459
8     cz 0.464
9     cz 0.722                                  $Fuel Outer Radius
10    cz 0.760                                  $Clad Outer Radius
*12   px 2.6971                                 $P/2
*13   px -2.6971
*14   py 2.6971
*15   py -2.6971
*16   pz 24.75                                  $H/2
*17   pz -24.75

mode:n
kcode 8000 1.0 50 450
ksrc 0.000 0.0 0.0
m1    8016.60c  4.6622E-02
      92234.60c 1.2799E-06
      92235.54c 1.6781E-04
      92238.54c 2.3137E-02                      $UO2
m2    8016.60c  4.6622E-02
      92234.60c 1.2799E-06
      92235.54c 1.6781E-04
      92238.54c 2.3137E-02                      $UO2
m3    8016.60c  4.6622E-02
      92234.60c 1.2799E-06
      92235.54c 1.6781E-04
      92238.54c 2.3137E-02                      $UO2
m4    8016.60c  4.6622E-02
      92234.60c 1.2799E-06
      92235.54c 1.6781E-04
      92238.54c 2.3137E-02                      $UO2
m5    8016.60c  4.6622E-02
      92234.60c 1.2799E-06
      92235.54c 1.6781E-04
      92238.54c 2.3137E-02                      $UO2
m6    8016.60c  4.6622E-02
      92234.60c 1.2799E-06
      92235.54c 1.6781E-04
      92238.54c 2.3137E-02                      $UO2
m7    8016.60c  4.6622E-02
```

```
      92234.60c 1.2799E-06
      92235.54c 1.6781E-04
      92238.54c 2.3137E-02                    $UO2
m8    8016.60c  4.6622E-02
      92234.60c 1.2799E-06
      92235.54c 1.6781E-04
      92238.54c 2.3137E-02                    $UO2
m9    8016.60c  4.6622E-02
      92234.60c 1.2799E-06
      92235.54c 1.6781E-04
      92238.54c 2.3137E-02                    $UO2
m10   24000.50c 7.5976E-05
      26000.55c 1.5562E-04
      40000.60c 4.2605E-02
      50000.35c 4.3269E-04                    $Zircaloy-4
m11   1001.60c  1.6640E-04
      1002.60c  6.6340E-02
      8016.60c  3.3250E-02                    $D2O
mt11  lwtr.02t
tmp   5.76E-08
      5.76E-08
      5.76E-08
      5.76E-08
      5.76E-08
      5.76E-08
      5.76E-08
      5.76E-08
      5.76E-08
      5.76E-08
      4.26E-08
      3.83E-08
      0.00E-00
      0.00E-00
      0.00E-00
      0.00E-00
      0.00E-00
      0.00E-00                                $Temperatures
xs1   35082.00c
      81.213094
      acebr82
      ~/Codes/MCNP/xsections
      1
      1
      1911
      0
      0
      2.585E-08
xs2   35083.00c
      82.202898
      acebr83
      ~/Codes/MCNP/xsections
      1
      1
      1911
      0
      0
      2.585E-08
xs3   36085.00c
      84.183100
      acekr85
      ~/Codes/MCNP/xsections
      1
      1
      2556
      0
      0
      2.585E-08
xs4   52131.00c
      129.781566
```

```
      acete131
      ~/Codes/MCNP/xsections
      1
      1
      1911
      0
      0
      2.585E-08
xs5   52132.00c
      130.775000
      acete132
      ~/Codes/MCNP/xsections
      1
      1
      2779
      0
      0
      2.585E-08
xs6   53130.00c
      128.791000
      acei130
      ~/Codes/MCNP/xsections
      1
      1
      2657
      0
      0
      2.585E-08
xs7   53131.00c
      129.782000
      acei131
      ~/Codes/MCNP/xsections
      1
      1
      2943
      0
      0
      2.585E-08
xs8   53132.00c
      130.774832
      acei132
      ~/Codes/MCNP/xsections
      1
      1
      1911
      0
      0
      2.585E-08
xs9   54130.00c
      128.788000
      acexe130
      ~/Codes/MCNP/xsections
      1
      1
      21096
      0
      0
      2.585E-08
xs10  54132.00c
      130.771000
      acexe132
      ~/Codes/MCNP/xsections
      1
      1
      9937
      0
      0
      2.585E-08
```

```
xs11 54136.00c
     134.740000
     acexe136
     ~/Codes/MCNP/xsections
     1
     1
     4429
     0
     0
     2.585E-08
print 40
```

```
Average CANDU-Nat.U Monteburns Input File
9
1
2
3
4
5
6
7
8
9
0.0
0.0
0.0
0.0
0.0
0.0
0.0
0.0
0.0
0.01524
-200.0
800
25
10
1
0
34
/nh/whirlpool/u/wscharl/Codes/Origen/Libs
0.005
0
28
28
28
28
28
28
28
28
28
28
28
28
28
28
28
28
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
```

```
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
```

```
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
```

```
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
```

```
54136.00c
92234.60c
92235.54c
92236.60c
92238.54c
93237.60c
93239.60c
94238.60c
94239.60c
94240.60c
94241.60c
94242.60c
35082.00c
35083.00c
36082.50c
36083.50c
36084.50c
36085.00c
36086.50c
52131.00c
52132.00c
53130.00c
53131.00c
53132.00c
54130.00c
54131.50c
54132.00c
54134.35c
54136.00c
```

APPENDIX C

Analysis Code Listing

```
Global Pvalue(0 To 20) As Double
Global pflag, Max, mflag As Integer
Global Model(1 To 20) As String
Global NumModels, ANumModels As Integer
Global Afile, Afile1, Afile2 As String
```

frmIntro

```
Option Explicit

Private Sub cmdAuthors_Click()
   frmIntro.Hide
   frmAuthors.Show
End Sub

Private Sub cmdExit_Click()
   Dim Message As String
   Dim ButtonsAndIcons As Integer
   Dim Title As String
   Dim Response As Integer

   Message = "Are you sure you want to quit?"
   ButtonsAndIcons = vbYesNo + vbQuestion
   Title = "Noble Gas Program"
   Response = MsgBox(Message, ButtonsAndIcons, Title)
   If Response = vbYes Then
      End
   End If
End Sub

Private Sub cmdOK_Click()
   Dim i As Integer
   pflag = 0
   mflag = 0
   Max = 20
   For i = 1 To Max
      Pvalue(i) = 1
   Next i
   frmIntro.Hide
   frmData.Show
End Sub
```

frmAuthors

```
Private Sub cmdOK_Click()
   frmIntro.Show
   frmAuthors.Hide
End Sub
```

frmData

```
Option Explicit

Private Sub chkDefault_Click()
   If chkDefault.value = 1 Then
      chkZeros.value = 0
      fraAir.Enabled = False
      txtA130.Text = 0.1553
      txtA131.Text = 0.803
      txtA132.Text = 1.0189
      txtA134.Text = 0.3939
      txtA136.Text = 0.3371
      txtA82.Text = 5.1556
      txtA83.Text = 5.1111
      txtA84.Text = 25.3333
      txtA85.Text = 0#
      txtA86.Text = 7.6889
      txtErrA130.Text = 0.000078
      txtErrA131.Text = 0.000402
      txtErrA132.Text = 0.000509
      txtErrA134.Text = 0.000197
      txtErrA136.Text = 0.000169
      txtErrA82.Text = 0.002578
      txtErrA83.Text = 0.002556
      txtErrA84.Text = 0.012667
      txtErrA85.Text = 0#
      txtErrA86.Text = 0.003844
   Else
      fraAir.Enabled = True
      txtA130.Text = ""
      txtA131.Text = ""
      txtA132.Text = ""
      txtA134.Text = ""
      txtA136.Text = ""
      txtA82.Text = ""
      txtA83.Text = ""
      txtA84.Text = ""
      txtA85.Text = ""
      txtA86.Text = ""
      txtErrA130.Text = ""
      txtErrA131.Text = ""
      txtErrA132.Text = ""
      txtErrA134.Text = ""
      txtErrA136.Text = ""
      txtErrA82.Text = ""
      txtErrA83.Text = ""
      txtErrA84.Text = ""
      txtErrA85.Text = ""
      txtErrA86.Text = ""
   End If
```

```
End Sub

Private Sub chkZeros_Click()
   If chkZeros.value = 1 Then
      chkDefault.value = 0
      fraAir.Enabled = False
      txtA130.Text = 0#
      txtA131.Text = 0#
      txtA132.Text = 0#
      txtA134.Text = 0#
      txtA136.Text = 0#
      txtA82.Text = 0#
      txtA83.Text = 0#
      txtA84.Text = 0#
      txtA85.Text = 0#
      txtA86.Text = 0#
      txtErrA130.Text = 0#
      txtErrA131.Text = 0#
      txtErrA132.Text = 0#
      txtErrA134.Text = 0#
      txtErrA136.Text = 0#
      txtErrA82.Text = 0#
      txtErrA83.Text = 0#
      txtErrA84.Text = 0#
      txtErrA85.Text = 0#
      txtErrA86.Text = 0#
   Else
      fraAir.Enabled = True
      txtA130.Text = ""
      txtA131.Text = ""
      txtA132.Text = ""
      txtA134.Text = ""
      txtA136.Text = ""
      txtA82.Text = ""
      txtA83.Text = ""
      txtA84.Text = ""
      txtA85.Text = ""
      txtA86.Text = ""
      txtErrA130.Text = ""
      txtErrA131.Text = ""
      txtErrA132.Text = ""
      txtErrA134.Text = ""
      txtErrA136.Text = ""
      txtErrA82.Text = ""
      txtErrA83.Text = ""
      txtErrA84.Text = ""
      txtErrA85.Text = ""
      txtErrA86.Text = ""
   End If
End Sub

Private Sub cmdAdvanced_Click()
```

```
    Afile1 = "Default"
    Afile2 = 1
    Afile = Afile1 + Afile2 + ".mdb"
    Unload frmAdvanced
    Load frmAdvanced
    frmIntro.Hide
    frmData.Hide
    frmAdvanced.Show
    frmSolution.Hide
    frmConfirm.Hide
End Sub

Private Sub cmdAnalyze_Click()

    frmConfirm.txtS130.Text = txtS130.Text
    frmConfirm.txtS131.Text = txtS131.Text
    frmConfirm.txtS132.Text = txtS132.Text
    frmConfirm.txtS134.Text = txtS134.Text
    frmConfirm.txtS136.Text = txtS136.Text
    frmConfirm.txtS82.Text = txtS82.Text
    frmConfirm.txtS83.Text = txtS83.Text
    frmConfirm.txtS84.Text = txtS84.Text
    frmConfirm.txtS85.Text = txtS85.Text
    frmConfirm.txtS86.Text = txtS86.Text
    frmConfirm.txtErrS130.Text = txtErrS130.Text
    frmConfirm.txtErrS131.Text = txtErrS131.Text
    frmConfirm.txtErrS132.Text = txtErrS132.Text
    frmConfirm.txtErrS134.Text = txtErrS134.Text
    frmConfirm.txtErrS136.Text = txtErrS136.Text
    frmConfirm.txtErrS82.Text = txtErrS82.Text
    frmConfirm.txtErrS83.Text = txtErrS83.Text
    frmConfirm.txtErrS84.Text = txtErrS84.Text
    frmConfirm.txtErrS85.Text = txtErrS85.Text
    frmConfirm.txtErrS86.Text = txtErrS86.Text

    frmConfirm.txtA130.Text = txtA130.Text
    frmConfirm.txtA131.Text = txtA131.Text
    frmConfirm.txtA132.Text = txtA132.Text
    frmConfirm.txtA134.Text = txtA134.Text
    frmConfirm.txtA136.Text = txtA136.Text
    frmConfirm.txtA82.Text = txtA82.Text
    frmConfirm.txtA83.Text = txtA83.Text
    frmConfirm.txtA84.Text = txtA84.Text
    frmConfirm.txtA85.Text = txtA85.Text
    frmConfirm.txtA86.Text = txtA86.Text
    frmConfirm.txtErrA130.Text = txtErrA130.Text
    frmConfirm.txtErrA131.Text = txtErrA131.Text
    frmConfirm.txtErrA132.Text = txtErrA132.Text
    frmConfirm.txtErrA134.Text = txtErrA134.Text
    frmConfirm.txtErrA136.Text = txtErrA136.Text
    frmConfirm.txtErrA82.Text = txtErrA82.Text
    frmConfirm.txtErrA83.Text = txtErrA83.Text
```

```
    frmConfirm.txtErrA84.Text = txtErrA84.Text
    frmConfirm.txtErrA85.Text = txtErrA85.Text
    frmConfirm.txtErrA86.Text = txtErrA86.Text

    frmIntro.Hide
    frmData.Hide
    frmAdvanced.Hide
    frmSolution.Hide
    frmConfirm.Show

End Sub

Private Sub cmdCancel_Click()
    txtS130.Text = ""
    txtS131.Text = ""
    txtS132.Text = ""
    txtS134.Text = ""
    txtS136.Text = ""
    txtS82.Text = ""
    txtS83.Text = ""
    txtS84.Text = ""
    txtS85.Text = ""
    txtS86.Text = ""
    txtA130.Text = ""
    txtA131.Text = ""
    txtA132.Text = ""
    txtA134.Text = ""
    txtA136.Text = ""
    txtA82.Text = ""
    txtA83.Text = ""
    txtA84.Text = ""
    txtA85.Text = ""
    txtA86.Text = ""
    txtErrS130.Text = ""
    txtErrS131.Text = ""
    txtErrS132.Text = ""
    txtErrS134.Text = ""
    txtErrS136.Text = ""
    txtErrS82.Text = ""
    txtErrS83.Text = ""
    txtErrS84.Text = ""
    txtErrS85.Text = ""
    txtErrS86.Text = ""
    txtErrA130.Text = ""
    txtErrA131.Text = ""
    txtErrA132.Text = ""
    txtErrA134.Text = ""
    txtErrA136.Text = ""
    txtErrA82.Text = ""
    txtErrA83.Text = ""
    txtErrA84.Text = ""
    txtErrA85.Text = ""
```

```
    txtErrA86.Text = ""
    chkDefault.value = 0
    frmIntro.Show
    frmData.Hide
    frmAdvanced.Hide
    frmSolution.Hide
    frmConfirm.Hide
End Sub

Private Sub Form_Load()
    Afile1 = "Default"
    Afile2 = "1"
    Afile = Afile1 + Afile2 + ".mdb"
End Sub
```

frmAdvanced

```
Option Explicit

Private Sub cmdCancel_Click()
    Dim i As Integer
    frmIntro.Hide
    frmData.Show
    frmAdvanced.Hide
    frmSolution.Hide
    frmConfirm.Hide
    pflag = 0
    For i = 1 To ANumModels
        Pvalue(i) = 1
    Next i
End Sub

Private Sub cmdOK_Click()
    Dim i, iflag As Integer

    Afile = Afile1 + Afile2 + ".mdb"

    iflag = 0
    For i = 1 To ANumModels
        If Pvalue(i) = 1 Then
            iflag = 1
        End If
    Next i
    If Pvalue(0) = 1 Then
        iflag = 0
    End If
    If iflag = 0 Then
        For i = 1 To ANumModels
            Pvalue(i) = 1
        Next i
    End If
```

```
    frmIntro.Hide
    frmData.Show
    frmAdvanced.Hide
    frmSolution.Hide
    frmConfirm.Hide
End Sub

Public Sub Form_Load()
    Dim Db As Database
    Dim Td As TableDef
    Dim Fd As Fields
    Dim i As Integer
    Dim dummy As String

    Afile = Afile1 + Afile2 + ".mdb"

    dummy = Afile1 + "2.mdb"
    Set Db = DBEngine.Workspaces(0).OpenDatabase(dummy)
    i = 0
    For Each Td In Db.TableDefs
        If (Td.Attributes And dbSystemObject) <> 0 Then
'           do nothing
        Else
            Set Fd = Td.Fields
            i = i + 1
            Model(i) = Td.Name
            frmAdvanced.lstForward.AddItem Format(Model(i) + " only")
        End If
    Next Td
    If Afile = dummy Then
        NumModels = i
        ANumModels = NumModels
    End If

    dummy = Afile1 + "1.mdb"
    Set Db = DBEngine.Workspaces(0).OpenDatabase(dummy)
    i = 0
    For Each Td In Db.TableDefs
        If (Td.Attributes And dbSystemObject) <> 0 Then
'           do nothing
        Else
            Set Fd = Td.Fields
            i = i + 1
            Model(i) = Td.Name
            frmAdvanced.lstBackward.AddItem Format(Model(i))
        End If
    Next Td
    If Afile = dummy Then
        NumModels = i
        ANumModels = NumModels
    End If
```

```
    For i = 1 To NumModels
      Pvalue(i) = 0
    Next i
    pflag = 0

    frmData.lblXe130.Enabled = True
    frmData.txtS130.Enabled = True
    frmData.txtErrS130.Enabled = True

    frmData.lblXe131.Enabled = True
    frmData.txtS131.Enabled = True
    frmData.txtErrS131.Enabled = True

    frmData.lblXe132.Enabled = True
    frmData.txtS132.Enabled = True
    frmData.txtErrS132.Enabled = True

    frmData.lblXe134.Enabled = True
    frmData.txtS134.Enabled = True
    frmData.txtErrS134.Enabled = True

    frmData.lblXe136.Enabled = True
    frmData.txtS136.Enabled = True
    frmData.txtErrS136.Enabled = True

    frmData.lblKr82.Enabled = True
    frmData.txtS82.Enabled = True
    frmData.txtErrS82.Enabled = True

    frmData.lblKr83.Enabled = True
    frmData.txtS83.Enabled = True
    frmData.txtErrS83.Enabled = True

    frmData.lblKr84.Enabled = True
    frmData.txtS84.Enabled = True
    frmData.txtErrS84.Enabled = True

    frmData.lblKr85.Enabled = True
    frmData.txtS85.Enabled = True
    frmData.txtErrS85.Enabled = True

    frmData.lblKr86.Enabled = True
    frmData.txtS86.Enabled = True
    frmData.txtErrS86.Enabled = True

End Sub

Private Sub lstBackward_ItemCheck(Item As Integer)
    Dim i As Integer
    pflag = 1
    For i = 0 To ANumModels
```

```
      If lstBackward.Selected(i) = False Then
        Pvalue(i) = 0
      Else
        Pvalue(i) = 1
      End If
    Next i
End Sub

Private Sub lstForward_ItemCheck(Item As Integer)
    Dim i As Integer
    pflag = 1
    For i = 0 To ANumModels - 1
      lstForward.Selected(i) = False
    Next i
    lstForward.Selected(Item) = True
    For i = 0 To 20
      Pvalue(i) = 0
    Next i
    If Pvalue(Item + 1) = 0 Then
      Pvalue(Item + 1) = 1
    Else
      Pvalue(Item + 1) = 0
    End If
End Sub


Private Sub optBackward_Click()
    Dim i As Integer
    Afile2 = "1"

'   For i = 0 To ANumModels - 1
'       lstForward.Selected(i) = False
'   Next i
    lblBackward.Enabled = True
    lstBackward.Enabled = True
    lblForward.Enabled = False
    lstForward.Enabled = False
    For i = 0 To ANumModels
      lstBackward.Selected(i) = False
    Next i
    mflag = 0
End Sub

Private Sub optForward_Click()
    Dim i As Integer
    Afile2 = "2"
    For i = 0 To ANumModels
      lstBackward.Selected(i) = False
    Next i
    lblBackward.Enabled = False
    lstBackward.Enabled = False
    lblForward.Enabled = True
```

```
    lstForward.Enabled = True
'   For i = 0 To ANumModels - 1
'       lstForward.Selected(i) = False
'   Next i
    mflag = 1
End Sub

Private Sub optHelios_Click()
    Dim i, j As Integer
    Afile1 = "Helios"
    For i = 1 To ANumModels
        j = ANumModels - i + 1
        frmAdvanced.lstBackward.RemoveItem (j)
        frmAdvanced.lstForward.RemoveItem (j - 1)
    Next i
    Form_Load

    frmData.lblXe130.Enabled = False
    frmData.txtS130.Enabled = False
    frmData.txtErrS130.Enabled = False
    frmData.txtS130.Text = " "
    frmData.txtErrS130.Text = " "

    frmData.lblXe131.Enabled = True
    frmData.txtS131.Enabled = True
    frmData.txtErrS131.Enabled = True

    frmData.lblXe132.Enabled = True
    frmData.txtS132.Enabled = True
    frmData.txtErrS132.Enabled = True

    frmData.lblXe134.Enabled = True
    frmData.txtS134.Enabled = True
    frmData.txtErrS134.Enabled = True

    frmData.lblXe136.Enabled = True
    frmData.txtS136.Enabled = True
    frmData.txtErrS136.Enabled = True

    frmData.lblKr82.Enabled = False
    frmData.txtS82.Enabled = False
    frmData.txtErrS82.Enabled = False
    frmData.txtS82.Text = " "
    frmData.txtErrS82.Text = " "

    frmData.lblKr83.Enabled = False
    frmData.txtS83.Enabled = False
    frmData.txtErrS83.Enabled = False
    frmData.txtS83.Text = " "
    frmData.txtErrS83.Text = " "

    frmData.lblKr84.Enabled = False
```

```
    frmData.txtS84.Enabled = False
    frmData.txtErrS84.Enabled = False
    frmData.txtS84.Text = " "
    frmData.txtErrS84.Text = " "

    frmData.lblKr85.Enabled = False
    frmData.txtS85.Enabled = False
    frmData.txtErrS85.Enabled = False
    frmData.txtS85.Text = " "
    frmData.txtErrS85.Text = " "

    frmData.lblKr86.Enabled = False
    frmData.txtS86.Enabled = False
    frmData.txtErrS86.Enabled = False
    frmData.txtS86.Text = " "
    frmData.txtErrS86.Text = " "

End Sub

Private Sub optMonteburns_Click()
    Dim i, j As Integer
    Afile1 = "Default"
    For i = 1 To ANumModels
        j = ANumModels - i + 1
        frmAdvanced.lstBackward.RemoveItem (j)
        frmAdvanced.lstForward.RemoveItem (j - 1)
    Next i
    Form_Load

    frmData.lblXe130.Enabled = True
    frmData.txtS130.Enabled = True
    frmData.txtErrS130.Enabled = True

    frmData.lblXe131.Enabled = True
    frmData.txtS131.Enabled = True
    frmData.txtErrS131.Enabled = True

    frmData.lblXe132.Enabled = True
    frmData.txtS132.Enabled = True
    frmData.txtErrS132.Enabled = True

    frmData.lblXe134.Enabled = True
    frmData.txtS134.Enabled = True
    frmData.txtErrS134.Enabled = True

    frmData.lblXe136.Enabled = True
    frmData.txtS136.Enabled = True
    frmData.txtErrS136.Enabled = True

    frmData.lblKr82.Enabled = True
    frmData.txtS82.Enabled = True
    frmData.txtErrS82.Enabled = True
```

```
    frmData.lblKr83.Enabled = True
    frmData.txtS83.Enabled = True
    frmData.txtErrS83.Enabled = True

    frmData.lblKr84.Enabled = True
    frmData.txtS84.Enabled = True
    frmData.txtErrS84.Enabled = True

    frmData.lblKr85.Enabled = True
    frmData.txtS85.Enabled = True
    frmData.txtErrS85.Enabled = True

    frmData.lblKr86.Enabled = True
    frmData.txtS86.Enabled = True
    frmData.txtErrS86.Enabled = True

End Sub

Private Sub optOrigen_Click()
    Dim i, j As Integer
    Afile1 = "Origen"
    For i = 1 To ANumModels
        j = ANumModels - i + 1
        frmAdvanced.lstBackward.RemoveItem (j)
        frmAdvanced.lstForward.RemoveItem (j - 1)
    Next i
    Form_Load

    frmData.lblXe130.Enabled = True
    frmData.txtS130.Enabled = True
    frmData.txtErrS130.Enabled = True

    frmData.lblXe131.Enabled = True
    frmData.txtS131.Enabled = True
    frmData.txtErrS131.Enabled = True

    frmData.lblXe132.Enabled = True
    frmData.txtS132.Enabled = True
    frmData.txtErrS132.Enabled = True

    frmData.lblXe134.Enabled = True
    frmData.txtS134.Enabled = True
    frmData.txtErrS134.Enabled = True

    frmData.lblXe136.Enabled = True
    frmData.txtS136.Enabled = True
    frmData.txtErrS136.Enabled = True

    frmData.lblKr82.Enabled = True
    frmData.txtS82.Enabled = True
    frmData.txtErrS82.Enabled = True
```

```
    frmData.lblKr83.Enabled = True
    frmData.txtS83.Enabled = True
    frmData.txtErrS83.Enabled = True

    frmData.lblKr84.Enabled = True
    frmData.txtS84.Enabled = True
    frmData.txtErrS84.Enabled = True

    frmData.lblKr85.Enabled = True
    frmData.txtS85.Enabled = True
    frmData.txtErrS85.Enabled = True

    frmData.lblKr86.Enabled = True
    frmData.txtS86.Enabled = True
    frmData.txtErrS86.Enabled = True

End Sub
```

frmConfirm

```
Option Explicit

Private Sub cmdCancel_Click()
    frmIntro.Hide
    frmData.Show
    frmAdvanced.Hide
    frmSolution.Hide
    frmConfirm.Hide
End Sub

Private Sub cmdConfirm_Click()
    Dim dummy As Double
    Dim i As Integer

    If pflag = 0 Then
        For i = 1 To 10
            Pvalue(i) = 1
        Next i
    End If

    'Calculate normalized and background corrected ratios and percent fission gases
    If Not IsNumeric(txtS130) Or Not IsNumeric(txtA130) Or Not IsNumeric(txtS134) Or Not
IsNumeric(txtA134) Or Not IsNumeric(txtErrS130) Or Not IsNumeric(txtErrA130) Or Not
IsNumeric(txtErrS134) Or Not IsNumeric(txtErrA134) Then
        frmSolution.txtXe130 = "not used"
        frmSolution.txtErrXe130 = "not used"
        frmSolution.txtPXe130 = "not used"
    Else
        dummy = (txtS130 - txtA130) / (txtS134 - txtA134)
        frmSolution.txtXe130 = Format$(dummy, "##0.0####")
```

```
    dummy = ((txtErrS130 - (-1# * txtErrA130)) / (txtS130 - txtA130)) ^ 2
    dummy = dummy + ((txtErrS134 - (-1# * txtErrA134)) / (txtS134 - txtA134)) ^ 2
    dummy = (txtS130 - txtA130) / (txtS134 - txtA134) * Sqr(dummy)
    frmSolution.txtErrXe130 = Format$(dummy, "##0.0####")
    dummy = (txtS130 - txtA130) / txtS130
    frmSolution.txtPXe130 = Format$(dummy, "##0.0## %")
    dummy = (txtS134 - txtA134) / txtS134
    frmSolution.txtPXe134 = Format$(dummy, "##0.0## %")
  End If
  If Not IsNumeric(txtS131) Or Not IsNumeric(txtA131) Or Not IsNumeric(txtS134) Or Not
IsNumeric(txtA134) Or Not IsNumeric(txtErrS131) Or Not IsNumeric(txtErrA131) Or Not
IsNumeric(txtErrS134) Or Not IsNumeric(txtErrA134) Then
    frmSolution.txtXe131 = "not used"
    frmSolution.txtErrXe131 = "not used"
    frmSolution.txtPXe131 = "not used"
  Else
    dummy = (txtS131 - txtA131) / (txtS134 - txtA134)
    frmSolution.txtXe131 = Format$(dummy, "##0.0####")
    dummy = ((txtErrS131 - (-1# * txtErrA131)) / (txtS131 - txtA131)) ^ 2
    dummy = dummy + ((txtErrS134 - (-1# * txtErrA134)) / (txtS134 - txtA134)) ^ 2
    dummy = (txtS131 - txtA131) / (txtS134 - txtA134) * Sqr(dummy)
    frmSolution.txtErrXe131 = Format$(dummy, "##0.0####")
    dummy = (txtS131 - txtA131) / txtS131
    frmSolution.txtPXe131 = Format$(dummy, "##0.0## %")
    dummy = (txtS134 - txtA134) / txtS134
    frmSolution.txtPXe134 = Format$(dummy, "##0.0## %")
  End If
  If Not IsNumeric(txtS132) Or Not IsNumeric(txtA132) Or Not IsNumeric(txtS134) Or Not
IsNumeric(txtA134) Or Not IsNumeric(txtErrS132) Or Not IsNumeric(txtErrA132) Or Not
IsNumeric(txtErrS134) Or Not IsNumeric(txtErrA134) Then
    frmSolution.txtXe132 = "not used"
    frmSolution.txtErrXe132 = "not used"
    frmSolution.txtPXe132 = "not used"
  Else
    dummy = (txtS132 - txtA132) / (txtS134 - txtA134)
    frmSolution.txtXe132 = Format$(dummy, "##0.0####")
    dummy = ((txtErrS132 - (-1# * txtErrA132)) / (txtS132 - txtA132)) ^ 2
    dummy = dummy + ((txtErrS134 - (-1# * txtErrA134)) / (txtS134 - txtA134)) ^ 2
    dummy = (txtS132 - txtA132) / (txtS134 - txtA134) * Sqr(dummy)
    frmSolution.txtErrXe132 = Format$(dummy, "##0.0####")
    dummy = (txtS132 - txtA132) / txtS132
    frmSolution.txtPXe132 = Format$(dummy, "##0.0## %")
    dummy = (txtS134 - txtA134) / txtS134
    frmSolution.txtPXe134 = Format$(dummy, "##0.0## %")
  End If
  If Not IsNumeric(txtS136) Or Not IsNumeric(txtA136) Or Not IsNumeric(txtS134) Or Not
IsNumeric(txtA134) Or Not IsNumeric(txtErrS136) Or Not IsNumeric(txtErrA136) Or Not
IsNumeric(txtErrS134) Or Not IsNumeric(txtErrA134) Then
    frmSolution.txtXe136 = "not used"
    frmSolution.txtErrXe136 = "not used"
    frmSolution.txtPXe136 = "not used"
  Else
```

```
    dummy = (txtS136 - txtA136) / (txtS134 - txtA134)
    frmSolution.txtXe136 = Format$(dummy, "##0.0####")
    dummy = ((txtErrS136 - (-1# * txtErrA136)) / (txtS136 - txtA136)) ^ 2
    dummy = dummy + ((txtErrS134 - (-1# * txtErrA134)) / (txtS134 - txtA134)) ^ 2
    dummy = (txtS136 - txtA136) / (txtS134 - txtA134) * Sqr(dummy)
    frmSolution.txtErrXe136 = Format$(dummy, "##0.0####")
    dummy = (txtS136 - txtA136) / txtS136
    frmSolution.txtPXe136 = Format$(dummy, "##0.0## %")
    dummy = (txtS134 - txtA134) / txtS134
    frmSolution.txtPXe134 = Format$(dummy, "##0.0## %")
  End If
  If Not IsNumeric(txtS82) Or Not IsNumeric(txtA82) Or Not IsNumeric(txtS86) Or Not
IsNumeric(txtA86) Or Not IsNumeric(txtErrS82) Or Not IsNumeric(txtErrA82) Or Not
IsNumeric(txtErrS86) Or Not IsNumeric(txtErrA86) Then
    frmSolution.txtKr82 = "not used"
    frmSolution.txtErrKr82 = "not used"
    frmSolution.txtPKr82 = "not used"
  Else
    dummy = (txtS82 - txtA82) / (txtS86 - txtA86)
    frmSolution.txtKr82 = Format$(dummy, "##0.0####")
    dummy = ((txtErrS82 - (-1# * txtErrA82)) / (txtS82 - txtA82)) ^ 2
    dummy = dummy + ((txtErrS86 - (-1# * txtErrA86)) / (txtS86 - txtA86)) ^ 2
    dummy = (txtS82 - txtA82) / (txtS86 - txtA86) * Sqr(dummy)
    frmSolution.txtErrKr82 = Format$(dummy, "##0.0####")
    dummy = (txtS82 - txtA82) / txtS82
    frmSolution.txtPKr82 = Format$(dummy, "##0.0## %")
    dummy = (txtS86 - txtA86) / txtS86
    frmSolution.txtPKr86 = Format$(dummy, "##0.0## %")
  End If
  If Not IsNumeric(txtS83) Or Not IsNumeric(txtA83) Or Not IsNumeric(txtS86) Or Not
IsNumeric(txtA86) Or Not IsNumeric(txtErrS83) Or Not IsNumeric(txtErrA83) Or Not
IsNumeric(txtErrS86) Or Not IsNumeric(txtErrA86) Then
    frmSolution.txtKr83 = "not used"
    frmSolution.txtErrKr83 = "not used"
    frmSolution.txtPKr83 = "not used"
  Else
    dummy = (txtS83 - txtA83) / (txtS86 - txtA86)
    frmSolution.txtKr83 = Format$(dummy, "##0.0####")
    dummy = ((txtErrS83 - (-1# * txtErrA83)) / (txtS83 - txtA83)) ^ 2
    dummy = dummy + ((txtErrS86 - (-1# * txtErrA86)) / (txtS86 - txtA86)) ^ 2
    dummy = (txtS83 - txtA83) / (txtS86 - txtA86) * Sqr(dummy)
    frmSolution.txtErrKr83 = Format$(dummy, "##0.0####")
    dummy = (txtS83 - txtA83) / txtS83
    frmSolution.txtPKr83 = Format$(dummy, "##0.0## %")
    dummy = (txtS86 - txtA86) / txtS86
    frmSolution.txtPKr86 = Format$(dummy, "##0.0## %")
  End If
  If Not IsNumeric(txtS84) Or Not IsNumeric(txtA84) Or Not IsNumeric(txtS86) Or Not
IsNumeric(txtA86) Or Not IsNumeric(txtErrS84) Or Not IsNumeric(txtErrA84) Or Not
IsNumeric(txtErrS86) Or Not IsNumeric(txtErrA86) Then
    frmSolution.txtKr84 = "not used"
    frmSolution.txtErrKr84 = "not used"
```

```
    frmSolution.txtPKr84 = "not used"
  Else
    dummy = (txtS84 - txtA84) / (txtS86 - txtA86)
    frmSolution.txtKr84 = Format$(dummy, "##0.0####")
    dummy = ((txtErrS84 - (-1# * txtErrA84)) / (txtS84 - txtA84)) ^ 2
    dummy = dummy + ((txtErrS86 - (-1# * txtErrA86)) / (txtS86 - txtA86)) ^ 2
    dummy = (txtS84 - txtA84) / (txtS86 - txtA86) * Sqr(dummy)
    frmSolution.txtErrKr84 = Format$(dummy, "##0.0####")
    dummy = (txtS84 - txtA84) / txtS84
    frmSolution.txtPKr84 = Format$(dummy, "##0.0## %")
    dummy = (txtS86 - txtA86) / txtS86
    frmSolution.txtPKr86 = Format$(dummy, "##0.0## %")
  End If
  If Not IsNumeric(txtS85) Or Not IsNumeric(txtA85) Or Not IsNumeric(txtS86) Or Not
IsNumeric(txtA86) Or Not IsNumeric(txtErrS85) Or Not IsNumeric(txtErrA85) Or Not
IsNumeric(txtErrS86) Or Not IsNumeric(txtErrA86) Then
    frmSolution.txtKr85 = "not used"
    frmSolution.txtErrKr85 = "not used"
    frmSolution.txtPKr85 = "not used"
  Else
    dummy = (txtS85 - txtA85) / (txtS86 - txtA86)
    frmSolution.txtKr85 = Format$(dummy, "##0.0####")
    dummy = ((txtErrS85 - (-1# * txtErrA85)) / (txtS85 - txtA85)) ^ 2
    dummy = dummy + ((txtErrS86 - (-1# * txtErrA86)) / (txtS86 - txtA86)) ^ 2
    dummy = (txtS85 - txtA85) / (txtS86 - txtA86) * Sqr(dummy)
    frmSolution.txtErrKr85 = Format$(dummy, "##0.0####")
    dummy = (txtS85 - txtA85) / txtS85
    frmSolution.txtPKr85 = Format$(dummy, "##0.0## %")
    dummy = (txtS86 - txtA86) / txtS86
    frmSolution.txtPKr86 = Format$(dummy, "##0.0## %")
  End If
  If Not IsNumeric(txtS86) Then
    frmSolution.txtPKr86 = "not used"
  ElseIf Not IsNumeric(txtS134) Then
    frmSolution.txtPXe136 = "not used"
  End If

' Display Computing Window
  Load frmCalculate
  frmIntro.Hide
  frmData.Hide
  frmAdvanced.Hide
  frmSolution.Hide
  frmConfirm.Hide
  frmCalculate.Show

End Sub
```

frmCalculate

```
Option Explicit
```

```
Dim pval, jflag As Integer

Private Sub Form_Load()
   Dim jflag As Integer
   FileDBase
   pval = 0
   frmSolution.Hide
End Sub


Public Sub FileDBase()

   Dim Db As Database
   Dim Td As TableDef
   Dim Fd As Fields
   Dim Rs As Recordset
   Dim Heading(1 To 31), eHeading(1 To 31), DumString As String
   Dim Age, ErrAge, TotAge As String
   Dim i, j, k, it, NumHeadings, iflag(1 To 50), Index As Integer
   Dim iset1, iset2, iset3, BNumModels As Integer
   Dim BurnInt(1 To 50, 1 To 9), Eint(1 To 50, 1 To 9), Point As Double
   Dim Dat1(1 To 31), eDat1(1 To 31), Dat2(1 To 31), eDat2(1 To 31) As Double
   Dim Data1, Data2, err1, err2, Burn1, Burn2, eplus, eminus As Double
   Dim Sum1, Sum2, Burn(1 To 50), Berror(1 To 50), ProductP As Double
   Dim Err, Rcalc(1 To 50, 1 To 31), eRcalc(1 To 50, 1 To 31) As Double
   Dim Pcalc(1 To 50, 1 To 9), PModel(0 To 50), R(1 To 9) As Double
   Dim SumP, Conf(1 To 50), dummy, halflife, N, No As Double
   Dim setpoint1, setpoint2, setpoint3, Emeas(1 To 9), value As Double
   Dim Norm(1 To 50), TotPu, ErrTotPu, FAge, thalf(1 To 31) As Double

' Zero Out Parameters
   For i = 1 To 50
      iflag(i) = -1
      Norm(i) = 1#
      For j = 1 To 9
         BurnInt(i, j) = 0#
         Eint(i, j) = 0#
      Next j
   Next i

' Set Heading Titles
   Heading(1) = "Xe-130/Xe-134"
   eHeading(1) = "errXe-130/Xe-134"
   Heading(2) = "Xe-131/Xe-134"
   eHeading(2) = "errXe-131/Xe-134"
   Heading(3) = "Xe-131/Xe-134"
   eHeading(3) = "errXe-131/Xe-134"
   Heading(4) = "Xe-132/Xe-134"
   eHeading(4) = "errXe-132/Xe-134"
   Heading(5) = "Xe-136/Xe-134"
   eHeading(5) = "errXe-136/Xe-134"
   Heading(6) = "Kr-82/Kr-86"
```

eHeading(6) = "errKr-82/Kr-86"
Heading(7) = "Kr-83/Kr-86"
eHeading(7) = "errKr-83/Kr-86"
Heading(8) = "Kr-84/Kr-86"
eHeading(8) = "errKr-84/Kr-86"
Heading(9) = "Kr-85/Kr-86"
eHeading(9) = "errKr-85/Kr-86"
Heading(10) = "Pu238"
eHeading(10) = "errPu238"
Heading(11) = "Pu239"
eHeading(11) = "errPu239"
Heading(12) = "Pu240"
eHeading(12) = "errPu240"
Heading(13) = "Pu241"
eHeading(13) = "errPu241"
Heading(14) = "Pu242"
eHeading(14) = "errPu242"
Heading(15) = "Pu240/239"
eHeading(15) = "errPu240/239"
Heading(16) = "Cs134"
eHeading(16) = "errCs134"
Heading(17) = "Cs135"
eHeading(17) = "errCs135"
Heading(18) = "Cs137"
eHeading(18) = "errCs137"
Heading(19) = "Tc99"
eHeading(19) = "errTc99"
Heading(20) = "I129"
eHeading(20) = "errI129"
Heading(21) = "Sr90"
eHeading(21) = "errSr90"
Heading(22) = "Nd146"
eHeading(22) = "errNd146"
Heading(23) = "Nd148"
eHeading(23) = "errNd148"
Heading(24) = "Np237"
eHeading(24) = "errNp237"
Heading(25) = "Am241"
eHeading(25) = "errAm241"
Heading(26) = "Am242m"
eHeading(26) = "errAm242m"
Heading(27) = "Am243"
eHeading(27) = "errAm243"
Heading(28) = "Cm242"
eHeading(28) = "errCm242"
Heading(29) = "Cm243"
eHeading(29) = "errCm243"
Heading(30) = "Cm244"
eHeading(30) = "errCm244"
Heading(31) = "Cm245"
eHeading(31) = "errCm245"

```
' Open Database and Get Model Names
   Set Db = DBEngine.Workspaces(0).OpenDatabase(Afile)
   i = 0
   For Each Td In Db.TableDefs
      If (Td.Attributes And dbSystemObject) <> 0 Then
'        do nothing
      Else
         Set Fd = Td.Fields
         i = i + 1
         Model(i) = Td.Name
      End If
   Next Td
   NumModels = i

'Scan Database and Interpolate for a Burnup for Each Ratio
   For i = 1 To NumModels

   Set Rs = Db.OpenRecordset(Model(i))
   If IsNumeric(frmSolution.txtXe130) Then
      Point = frmSolution.txtXe130
      Err = frmSolution.txtErrXe130
      Emeas(1) = Err
      Rs.MoveFirst
      Rs.MoveNext
      Do Until Rs.EOF
         Rs.MovePrevious
         Data1 = 1# * Rs("Xe-130/Xe-134")
         err1 = 1# * Rs("errXe-130/Xe-134")
         Burn1 = 1# * Rs("Burnup")
         Rs.MoveNext
         Data2 = 1# * Rs("Xe-130/Xe-134")
         err2 = 1# * Rs("errXe-130/Xe-134")
         Burn2 = 1# * Rs("Burnup")
         If Point <= Data1 And Point > Data2 Then
            BurnInt(i, 1) = Burn1 + ((Point - Data1) / ((Data2 - Data1) / (Burn2 - Burn1)))
            eplus = (Point - Err - Data1 - err1) * (Burn2 - Burn1)
            eplus = eplus / (Data2 + err2 - Data1 - err1)
            eplus = eplus + Burn1 - BurnInt(i, 1)
            eminus = (Point + Err - Data1 + err1) * (Burn2 - Burn1)
            eminus = eminus / (Data2 - err2 - Data1 + err1)
            eminus = BurnInt(i, 1) - Burn1 - eminus
            Eint(i, 1) = (eplus + eminus) / 2#
         ElseIf Point >= Data1 And Point < Data2 Then
            BurnInt(i, 1) = Burn1 + ((Point - Data1) / ((Data2 - Data1) / (Burn2 - Burn1)))
            eplus = (Point + Err - Data1 + err1) * (Burn2 - Burn1)
            eplus = eplus / (Data2 - err2 - Data1 + err1)
            eplus = eplus + Burn1 - BurnInt(i, 1)
            eminus = (Point - Err - Data1 - err1) * (Burn2 - Burn1)
            eminus = eminus / (Data2 + err2 - Data1 - err1)
            eminus = BurnInt(i, 1) - Burn1 - eminus
            Eint(i, 1) = (eplus + eminus) / 2#
         End If
```

```
      Rs.MoveNext
    Loop
  End If
  If IsNumeric(frmSolution.txtXe131) Then
    Point = frmSolution.txtXe131
    Err = frmSolution.txtErrXe131
    Emeas(2) = Err
    Rs.MoveFirst
    Rs.MoveNext
    Do Until Rs.EOF
      Rs.MovePrevious
      Data1 = 1# * Rs("Xe-131/Xe-134")
      err1 = 1# * Rs("errXe-131/Xe-134")
      Burn1 = 1# * Rs("Burnup")
      Rs.MoveNext
      Data2 = 1# * Rs("Xe-131/Xe-134")
      err2 = 1# * Rs("errXe-131/Xe-134")
      Burn2 = 1# * Rs("Burnup")
      If Point <= Data1 And Point > Data2 Then
        BurnInt(i, 2) = Burn1 + ((Point - Data1) / ((Data2 - Data1) / (Burn2 - Burn1)))
        eplus = (Point - Err - Data1 - err1) * (Burn2 - Burn1)
        eplus = eplus / (Data2 + err2 - Data1 - err1)
        eplus = eplus + Burn1 - BurnInt(i, 2)
        eminus = (Point + Err - Data1 + err1) * (Burn2 - Burn1)
        eminus = eminus / (Data2 - err2 - Data1 + err1)
        eminus = BurnInt(i, 2) - Burn1 - eminus
        Eint(i, 2) = (eplus + eminus) / 2#
      ElseIf Point >= Data1 And Point < Data2 Then
        BurnInt(i, 3) = Burn1 + ((Point - Data1) / ((Data2 - Data1) / (Burn2 - Burn1)))
        eplus = (Point + Err - Data1 + err1) * (Burn2 - Burn1)
        eplus = eplus / (Data2 - err2 - Data1 + err1)
        eplus = eplus + Burn1 - BurnInt(i, 3)
        eminus = (Point - Err - Data1 - err1) * (Burn2 - Burn1)
        eminus = eminus / (Data2 + err2 - Data1 - err1)
        eminus = BurnInt(i, 3) - Burn1 - eminus
        Eint(i, 3) = (eplus + eminus) / 2#
      End If
      Rs.MoveNext
    Loop
  End If
  If IsNumeric(frmSolution.txtXe132) Then
    Point = frmSolution.txtXe132
    Err = frmSolution.txtErrXe132
    Emeas(4) = Err
    Rs.MoveFirst
    Rs.MoveNext
    Do Until Rs.EOF
      Rs.MovePrevious
      Data1 = 1# * Rs("Xe-132/Xe-134")
      err1 = 1# * Rs("errXe-132/Xe-134")
      Burn1 = 1# * Rs("Burnup")
      Rs.MoveNext
```

```
      Data2 = 1# * Rs("Xe-132/Xe-134")
      err2 = 1# * Rs("errXe-132/Xe-134")
      Burn2 = 1# * Rs("Burnup")
      If Point <= Data1 And Point > Data2 Then
        BurnInt(i, 4) = Burn1 + ((Point - Data1) / ((Data2 - Data1) / (Burn2 - Burn1)))
        eplus = (Point - Err - Data1 - err1) * (Burn2 - Burn1)
        eplus = eplus / (Data2 + err2 - Data1 - err1)
        eplus = eplus + Burn1 - BurnInt(i, 4)
        eminus = (Point + Err - Data1 + err1) * (Burn2 - Burn1)
        eminus = eminus / (Data2 - err2 - Data1 + err1)
        eminus = BurnInt(i, 4) - Burn1 - eminus
        Eint(i, 4) = (eplus + eminus) / 2#
      ElseIf Point >= Data1 And Point < Data2 Then
        BurnInt(i, 4) = Burn1 + ((Point - Data1) / ((Data2 - Data1) / (Burn2 - Burn1)))
        eplus = (Point + Err - Data1 + err1) * (Burn2 - Burn1)
        eplus = eplus / (Data2 - err2 - Data1 + err1)
        eplus = eplus + Burn1 - BurnInt(i, 4)
        eminus = (Point - Err - Data1 - err1) * (Burn2 - Burn1)
        eminus = eminus / (Data2 + err2 - Data1 - err1)
        eminus = BurnInt(i, 4) - Burn1 - eminus
        Eint(i, 4) = (eplus + eminus) / 2#
      End If
      Rs.MoveNext
    Loop
End If
If IsNumeric(frmSolution.txtKr82) Then
    Point = frmSolution.txtKr82
    Err = frmSolution.txtErrKr82
    Emeas(6) = Err
    Rs.MoveFirst
    Rs.MoveNext
    Do Until Rs.EOF
      Rs.MovePrevious
      Data1 = 1# * Rs("Kr-82/Kr-86")
      err1 = 1# * Rs("errKr-82/Kr-86")
      Burn1 = 1# * Rs("Burnup")
      Rs.MoveNext
      Data2 = 1# * Rs("Kr-82/Kr-86")
      err2 = 1# * Rs("errKr-82/Kr-86")
      Burn2 = 1# * Rs("Burnup")
      If Point <= Data1 And Point > Data2 Then
        BurnInt(i, 6) = Burn1 + ((Point - Data1) / ((Data2 - Data1) / (Burn2 - Burn1)))
        eplus = (Point - Err - Data1 - err1) * (Burn2 - Burn1)
        eplus = eplus / (Data2 + err2 - Data1 - err1)
        eplus = eplus + Burn1 - BurnInt(i, 6)
        eminus = (Point + Err - Data1 + err1) * (Burn2 - Burn1)
        eminus = eminus / (Data2 - err2 - Data1 + err1)
        eminus = BurnInt(i, 6) - Burn1 - eminus
        Eint(i, 6) = (eplus + eminus) / 2#
      ElseIf Point >= Data1 And Point < Data2 Then
        BurnInt(i, 6) = Burn1 + ((Point - Data1) / ((Data2 - Data1) / (Burn2 - Burn1)))
        eplus = (Point + Err - Data1 + err1) * (Burn2 - Burn1)
```

```
            eplus = eplus / (Data2 - err2 - Data1 + err1)
            eplus = eplus + Burn1 - BurnInt(i, 6)
            eminus = (Point - Err - Data1 - err1) * (Burn2 - Burn1)
            eminus = eminus / (Data2 + err2 - Data1 - err1)
            eminus = BurnInt(i, 6) - Burn1 - eminus
            Eint(i, 6) = (eplus + eminus) / 2#
        End If
        Rs.MoveNext
    Loop
End If
If IsNumeric(frmSolution.txtKr83) Then
    Point = frmSolution.txtKr83
    Err = frmSolution.txtErrKr83
    Emeas(7) = Err
    Rs.MoveFirst
    Rs.MoveNext
    Do Until Rs.EOF
        Rs.MovePrevious
        Data1 = 1# * Rs("Kr-83/Kr-86")
        err1 = 1# * Rs("errKr-83/Kr-86")
        Burn1 = 1# * Rs("Burnup")
        Rs.MoveNext
        Data2 = 1# * Rs("Kr-83/Kr-86")
        err2 = 1# * Rs("errKr-83/Kr-86")
        Burn2 = 1# * Rs("Burnup")
        If Point <= Data1 And Point > Data2 Then
            BurnInt(i, 7) = Burn1 + ((Point - Data1) / ((Data2 - Data1) / (Burn2 - Burn1)))
            eplus = (Point - Err - Data1 - err1) * (Burn2 - Burn1)
            eplus = eplus / (Data2 + err2 - Data1 - err1)
            eplus = eplus + Burn1 - BurnInt(i, 7)
            eminus = (Point + Err - Data1 + err1) * (Burn2 - Burn1)
            eminus = eminus / (Data2 - err2 - Data1 + err1)
            eminus = BurnInt(i, 7) - Burn1 - eminus
            Eint(i, 7) = (eplus + eminus) / 2#
        ElseIf Point >= Data1 And Point < Data2 Then
            BurnInt(i, 7) = Burn1 + ((Point - Data1) / ((Data2 - Data1) / (Burn2 - Burn1)))
            eplus = (Point + Err - Data1 + err1) * (Burn2 - Burn1)
            eplus = eplus / (Data2 - err2 - Data1 + err1)
            eplus = eplus + Burn1 - BurnInt(i, 7)
            eminus = (Point - Err - Data1 - err1) * (Burn2 - Burn1)
            eminus = eminus / (Data2 + err2 - Data1 - err1)
            eminus = BurnInt(i, 7) - Burn1 - eminus
            Eint(i, 7) = (eplus + eminus) / 2#
        End If
        Rs.MoveNext
    Loop
End If
If IsNumeric(frmSolution.txtKr84) Then
    Point = frmSolution.txtKr84
    Err = frmSolution.txtErrKr84
    Emeas(8) = Err
    Rs.MoveFirst
```

```
    Rs.MoveNext
    Do Until Rs.EOF
      Rs.MovePrevious
      Data1 = 1# * Rs("Kr-84/Kr-86")
      err1 = 1# * Rs("errKr-84/Kr-86")
      Burn1 = 1# * Rs("Burnup")
      Rs.MoveNext
      Data2 = 1# * Rs("Kr-84/Kr-86")
      err2 = 1# * Rs("errKr-84/Kr-86")
      Burn2 = 1# * Rs("Burnup")
      If Point <= Data1 And Point > Data2 Then
        BurnInt(i, 8) = Burn1 + ((Point - Data1) / ((Data2 - Data1) / (Burn2 - Burn1)))
        eplus = (Point - Err - Data1 - err1) * (Burn2 - Burn1)
        eplus = eplus / (Data2 + err2 - Data1 - err1)
        eplus = eplus + Burn1 - BurnInt(i, 8)
        eminus = (Point + Err - Data1 + err1) * (Burn2 - Burn1)
        eminus = eminus / (Data2 - err2 - Data1 + err1)
        eminus = BurnInt(i, 8) - Burn1 - eminus
        Eint(i, 8) = (eplus + eminus) / 2#
      ElseIf Point >= Data1 And Point < Data2 Then
        BurnInt(i, 8) = Burn1 + ((Point - Data1) / ((Data2 - Data1) / (Burn2 - Burn1)))
        eplus = (Point + Err - Data1 + err1) * (Burn2 - Burn1)
        eplus = eplus / (Data2 - err2 - Data1 + err1)
        eplus = eplus + Burn1 - BurnInt(i, 8)
        eminus = (Point - Err - Data1 - err1) * (Burn2 - Burn1)
        eminus = eminus / (Data2 + err2 - Data1 - err1)
        eminus = BurnInt(i, 8) - Burn1 - eminus
        Eint(i, 8) = (eplus + eminus) / 2#
      End If
      Rs.MoveNext
    Loop
  End If

  Next i

' Check for Non-Monotonic Models
  For i = 1 To NumModels
    If BurnInt(i, 2) = 0 And BurnInt(i, 3) <> 0 Then
      BurnInt(i, 2) = BurnInt(i, 3)
      Eint(i, 2) = Eint(i, 3)
      BurnInt(i, 3) = 0#
      Eint(i, 3) = 0#
    ElseIf BurnInt(i, 2) <> 0 And BurnInt(i, 3) <> 0 Then
      BurnInt(NumModels + 1, 1) = BurnInt(i, 1)
      Eint(NumModels + 1, 1) = Eint(i, 1)
      BurnInt(NumModels + 1, 2) = BurnInt(i, 3)
      Eint(NumModels + 1, 2) = Eint(i, 3)
      For j = 4 To 9
        BurnInt(NumModels + 1, j) = BurnInt(i, j)
        Eint(NumModels + 1, j) = Eint(i, j)
      Next j
      BurnInt(i, 3) = 0#
```

```
        Eint(i, 3) = 0#
        NumModels = NumModels + 1
        Model(NumModels) = Model(i)
        Pvalue(NumModels) = Pvalue(i)
      End If
    Next i

' Set Flag for Useable Model Solutions
    For i = 1 To NumModels
      For j = 1 To 9
        If BurnInt(i, j) <> 0 Then
          iflag(i) = iflag(i) + 1
        End If
      Next j
    Next i

' Check if Any Useable Models were Entered
    jflag = 0
    For i = 1 To NumModels
      If iflag(i) > 0 Then
        jflag = 1
      End If
    Next i
    If jflag = 0 Then
      Dim Message, Title As String
      Dim ButtonsAndIcons As Integer
      Message = "No selected reactor types fit the input ratios!"
      ButtonsAndIcons = vbOKOnly + vbExclamation
      Title = "Error in Input Ratios!"
      MsgBox Message, ButtonsAndIcons, Title
      Exit Sub
    End If

' Eliminate Unuseable Models
    it = 0
    BNumModels = NumModels
    For i = 1 To NumModels
      it = it + 1
      If iflag(it) < 1 Then
        For j = 1 To BNumModels - it
          Model(it + j - 1) = Model(it + j)
          iflag(it + j - 1) = iflag(it + j)
          Pvalue(it + j - 1) = Pvalue(it + j)
          For k = 1 To 9
            BurnInt(it + j - 1, k) = BurnInt(it + j, k)
            Eint(it + j - 1, k) = Eint(it + j, k)
          Next k
        Next j
        BNumModels = BNumModels - 1
        it = it - 1
      End If
    Next i
```

```
NumModels = BNumModels

' Generate a Combined Average Burnup and Error for Each Model
For i = 1 To NumModels
   Sum1 = 0#
   Sum2 = 0#
   For j = 1 To 9
      If BurnInt(i, j) <> 0 Then
         Sum1 = Sum1 + BurnInt(i, j) / Eint(i, j) / Eint(i, j)
         Sum2 = Sum2 + (1# / Eint(i, j) / Eint(i, j))
      End If
   Next j
   Burn(i) = Sum1 / Sum2
   Berror(i) = Sqr(1# / Sum2)
Next i

' Generate Ratios from Database Based on Combined Average Burnup
setpoint1 = 0#
setpoint2 = 0#
setpoint3 = 0#
iset1 = 0
iset2 = 0
iset1 = 0
SumP = 0#
For i = 1 To NumModels
   Set Rs = Db.OpenRecordset(Model(i))
   Rs.MoveFirst
   Rs.MoveNext
   Do Until Rs.EOF
      Rs.MovePrevious
      For j = 1 To 31
         Dat1(j) = Rs(Heading(j))
         eDat1(j) = Rs(eHeading(j))
      Next j
      Burn1 = Rs("Burnup")
      Rs.MoveNext
      For j = 1 To 31
         Dat2(j) = Rs(Heading(j))
         eDat2(j) = Rs(eHeading(j))
      Next j
      Burn2 = Rs("Burnup")
      If Burn(i) > Burn1 And Burn(i) < Burn2 Then
         For j = 1 To 31
         If Dat1(j) > Dat2(j) Then
            Rcalc(i, j) = (Burn(i) - Burn1) * (Dat2(j) - Dat1(j))
            Rcalc(i, j) = (Rcalc(i, j) / (Burn2 - Burn1)) + Dat1(j)
            eplus = (Burn(i) - Berror(i) - Burn1)
            eplus = eplus * (Dat2(j) + eDat2(j) - Dat1(j) - eDat1(j))
            eplus = eplus / (Burn2 - Burn1)
            eplus = eplus + Dat1(j) + eDat1(j) - Rcalc(i, j)
            eminus = (Burn(i) + Berror(i) - Burn1)
            eminus = eminus * (Dat2(j) - eDat2(j) - Dat1(j) + eDat1(j))
```

```
            eminus = eminus / (Burn2 - Burn1)
            eminus = Rcalc(i, j) - Dat1(j) + eDat1(j) - eminus
            eRcalc(i, j) = (eplus + eminus) / 2#
          Else
            Rcalc(i, j) = (Burn(i) - Burn1) * (Dat2(j) - Dat1(j))
            Rcalc(i, j) = (Rcalc(i, j) / (Burn2 - Burn1)) + Dat1(j)
            eplus = (Burn(i) + Berror(i) - Burn1)
            eplus = eplus * (Dat2(j) + eDat2(j) - Dat1(j) - eDat1(j))
            eplus = eplus / (Burn2 - Burn1)
            eplus = eplus + Dat1(j) + eDat1(j) - Rcalc(i, j)
            eminus = (Burn(i) - Berror(i) - Burn1)
            eminus = eminus * (Dat2(j) - eDat2(j) - Dat1(j) + eDat1(j))
            eminus = eminus / (Burn2 - Burn1)
            eminus = Rcalc(i, j) - Dat1(j) + eDat1(j) - eminus
            eRcalc(i, j) = (eplus + eminus) / 2#
          End If
          Next j
        End If
        Rs.MoveNext
      Loop

' Calculate Model Based Probabilities for Each Ratio
      If IsNumeric(frmSolution.txtXe130) Then
        R(1) = frmSolution.txtXe130
      Else
        R(1) = 0#
      End If
      If IsNumeric(frmSolution.txtXe131) Then
        R(2) = frmSolution.txtXe131
      Else
        R(2) = 0#
      End If
      If IsNumeric(frmSolution.txtXe131) Then
        R(3) = frmSolution.txtXe131
      Else
        R(3) = 0#
      End If
      If IsNumeric(frmSolution.txtXe132) Then
        R(4) = frmSolution.txtXe132
      Else
        R(4) = 0#
      End If
      If IsNumeric(frmSolution.txtXe136) Then
        R(5) = frmSolution.txtXe130
      Else
        R(5) = 0#
      End If
      If IsNumeric(frmSolution.txtKr82) Then
        R(6) = frmSolution.txtKr82
      Else
        R(6) = 0#
      End If
```

```
    If IsNumeric(frmSolution.txtKr83) Then
      R(7) = frmSolution.txtKr83
    Else
      R(7) = 0#
    End If
    If IsNumeric(frmSolution.txtKr84) Then
      R(8) = frmSolution.txtKr84
    Else
      R(8) = 0#
    End If
    If IsNumeric(frmSolution.txtKr85) Then
      R(9) = frmSolution.txtKr85
    Else
      R(9) = 0#
    End If
    For j = 1 To 9
      If R(j) > 0# Then
        If BurnInt(i, j) <> 0# Then
          value = 1# / ((2# * 3.1415926) ^ 0.5) / eRcalc(i, j)
          Norm(i) = Norm(i) * value
          Pcalc(i, j) = (R(j) - Rcalc(i, j)) * (R(j) - Rcalc(i, j))
          Pcalc(i, j) = Pcalc(i, j) / 2# / eRcalc(i, j) / eRcalc(i, j)
          Pcalc(i, j) = value * Exp(-Pcalc(i, j))
        Else
          Pcalc(i, j) = 0#
        End If
      Else
        Pcalc(i, 1) = 1#
      End If
    Next j
    Pcalc(i, 3) = 1#
  Next i

' Generate Comparison Probabilities Using Bayes Therorem
  For i = 1 To NumModels
    ProductP = 1#
    If IsNumeric(frmSolution.txtXe130) Then
      ProductP = ProductP * Pcalc(i, 1)
    End If
    If IsNumeric(frmSolution.txtXe131) Then
      ProductP = ProductP * Pcalc(i, 2)
    End If
    If IsNumeric(frmSolution.txtXe132) Then
      ProductP = ProductP * Pcalc(i, 4)
    End If
    If IsNumeric(frmSolution.txtKr82) Then
      ProductP = ProductP * Pcalc(i, 6)
    End If
    If IsNumeric(frmSolution.txtKr83) Then
      ProductP = ProductP * Pcalc(i, 7)
    End If
    If IsNumeric(frmSolution.txtKr84) Then
```

```
        ProductP = ProductP * Pcalc(i, 8)
      End If
      PModel(i) = Pvalue(i) * ProductP / Norm(i)

' Sort for Best Model
      If PModel(i) > setpoint1 Then
        setpoint3 = setpoint2
        iset3 = iset2
        setpoint2 = setpoint1
        iset2 = iset1
        setpoint1 = PModel(i)
        iset1 = i
      End If
      If PModel(i) < setpoint1 And PModel(i) > setpoint2 Then
        setpoint3 = setpoint2
        iset3 = iset2
        setpoint2 = PModel(i)
        iset2 = i
      End If
      If PModel(i) < setpoint2 And PModel(i) > setpoint3 Then
        setpoint3 = PModel(i)
        iset3 = i
      End If
      SumP = SumP + PModel(i)

    Next i

    If iset1 = 0 Then
      Message = "No selected reactor types fit the input ratios!"
      ButtonsAndIcons = vbOKOnly + vbExclamation
      Title = "Error in Input Ratios!"
      MsgBox Message, ButtonsAndIcons, Title
      jflag = 0
      Exit Sub
    End If

' Set Confidence Level for Each Model
    If mflag = 0 Then
      For i = 1 To NumModels
        Conf(i) = 0.9999 * PModel(i) / SumP
      Next i
    Else
      For i = 1 To NumModels
        Conf(i) = PModel(i)
      Next i
    End If

' Calculate and Output Fuel Age
    If IsNumeric(frmSolution.txtKr85) And IsNumeric(frmSolution.txtErrKr85) Then
      halflife = 10.73
      N = frmSolution.txtKr85
      No = Rcalc(iset1, 9)
```

```
    FAge = (150# / 365#) - (halflife * Log(N / No) / Log(2#))
    frmSolution.txtAge = Format$(FAge, "0.000")
    Age = frmSolution.txtAge
    dummy = frmSolution.txtErrKr85
    N = 1# * N + 1# * dummy
    No = Rcalc(iset1, 9) - eRcalc(iset1, 9)
    dummy = -halflife * Log(N / No) / Log(2#)
    dummy = Age - dummy
    frmSolution.txtAge = Format$(dummy, "0.000")
    ErrAge = frmSolution.txtAge
    TotAge = Age + " +/- " + ErrAge
    frmSolution.txtAge = TotAge
  Else
    frmSolution.txtAge = "not calculated"
  End If

' Calculate and Output Fuel Operating History
  If IsNumeric(frmSolution.txtXe136) Then
'   do something
  Else
    frmSolution.txtHistory = "not calculated"
  End If


' Output Best Model Solution
  frmSolution.txtType = Model(iset1)
  frmSolution.txtBurnup = Format$(Burn(iset1), "####00.00")
  frmSolution.txtError = Format$(Berror(iset1), "####00.00")
  frmSolution.txtConfidence = Format$(Conf(iset1), "0.000 %")

' Output Secondary Solutions
  frmSolution.txtType1 = Model(iset1)
  frmSolution.txtBurn1 = Format$(Burn(iset1), "####00.00")
  frmSolution.txtConf1 = Format$(Conf(iset1), "0.000 %")
  If iset2 > 0 Then
    frmSolution.txtType2 = Model(iset2)
    frmSolution.txtBurn2 = Format$(Burn(iset2), "####00.00")
    frmSolution.txtConf2 = Format$(Conf(iset2), "0.000 %")
  Else
    frmSolution.txtType2 = "not used"
    frmSolution.txtBurn2 = "not used"
    frmSolution.txtConf2 = "not used"
  End If
  If iset3 > 0 Then
    frmSolution.txtType3 = Model(iset3)
    frmSolution.txtBurn3 = Format$(Burn(iset3), "####00.00")
    frmSolution.txtConf3 = Format$(Conf(iset3), "0.000 %")
  Else
    frmSolution.txtType3 = "not used"
    frmSolution.txtBurn3 = "not used"
    frmSolution.txtConf3 = "not used"
  End If
```

```
' Display Assembly Picture for Best Model
  Dim FileNum As Integer
  If Model(iset1) = "US Production Reactor Target Fuel" Then
     Set frmSolution.pctFuel = LoadPicture("USP.gif")
     FileNum = FreeFile
     Open "USP.txt" For Input As #FileNum
     Do While Not EOF(FileNum)
        Input #FileNum, DumString
        frmSolution.lstModels.AddItem DumString
     Loop
     Close #FileNum
  End If
  If Model(iset1) = "PWR" Then
     Set frmSolution.pctFuel = LoadPicture("PWR.bmp")
     FileNum = FreeFile
     Open "PWR.txt" For Input As #FileNum
     Do While Not EOF(FileNum)
        Input #FileNum, DumString
        frmSolution.lstModels.AddItem DumString
     Loop
     Close #FileNum
  End If
  If Model(iset1) = "BWR" Then
     Set frmSolution.pctFuel = LoadPicture("BWR.bmp")
     FileNum = FreeFile
     Open "BWR.txt" For Input As #FileNum
     Do While Not EOF(FileNum)
        Input #FileNum, DumString
        frmSolution.lstModels.AddItem DumString
     Loop
     Close #FileNum
  End If
  If Model(iset1) = "CANDU" Then
     Set frmSolution.pctFuel = LoadPicture("CANDU.bmp")
     FileNum = FreeFile
     Open "CANDU.txt" For Input As #FileNum
     Do While Not EOF(FileNum)
        Input #FileNum, DumString
        frmSolution.lstModels.AddItem DumString
     Loop
     Close #FileNum
  End If
  If Model(iset1) = "RBMK" Then
     Set frmSolution.pctFuel = LoadPicture("RBMK.bmp")
     FileNum = FreeFile
     Open "RBMK.txt" For Input As #FileNum
     Do While Not EOF(FileNum)
        Input #FileNum, DumString
        frmSolution.lstModels.AddItem DumString
     Loop
     Close #FileNum
```

```
End If
If Model(iset1) = "HTGR" Then
   Set frmSolution.pctFuel = LoadPicture("HTGR.bmp")
   FileNum = FreeFile
   Open "HTGR.txt" For Input As #FileNum
   Do While Not EOF(FileNum)
      Input #FileNum, DumString
      frmSolution.lstModels.AddItem DumString
   Loop
   Close #FileNum
End If
If Model(iset1) = "LMFBR Driver Fuel" Then
   Set frmSolution.pctFuel = LoadPicture("LMFBR.bmp")
   FileNum = FreeFile
   Open "LMFBR.txt" For Input As #FileNum
   Do While Not EOF(FileNum)
      Input #FileNum, DumString
      frmSolution.lstModels.AddItem DumString
   Loop
   Close #FileNum
End If
If Model(iset1) = "LMFBR Blanket Fuel" Then
   Set frmSolution.pctFuel = LoadPicture("LMFBR.bmp")
   FileNum = FreeFile
   Open "LMFBR.txt" For Input As #FileNum
   Do While Not EOF(FileNum)
      Input #FileNum, DumString
      frmSolution.lstModels.AddItem DumString
   Loop
   Close #FileNum
End If
If Model(iset1) = "GE BWR/6" Then
   Set frmSolution.pctFuel = LoadPicture("GE-BWR6.bmp")
   FileNum = FreeFile
   Open "BWR.txt" For Input As #FileNum
   Do While Not EOF(FileNum)
      Input #FileNum, DumString
      frmSolution.lstModels.AddItem DumString
   Loop
   Close #FileNum
End If
If Model(iset1) = "Westinghouse 17x17 PWR" Then
   Set frmSolution.pctFuel = LoadPicture("Westinghouse 17x17 PWR.bmp")
   FileNum = FreeFile
   Open "PWR.txt" For Input As #FileNum
   Do While Not EOF(FileNum)
      Input #FileNum, DumString
      frmSolution.lstModels.AddItem DumString
   Loop
   Close #FileNum
End If
```

```
' Output Calculated and Measured Ratios for Best Solution
   frmAddition.txtMRatio(0) = frmSolution.txtXe130
   frmAddition.txtMRatio(1) = frmSolution.txtXe131
   frmAddition.txtMRatio(2) = frmSolution.txtXe132
   frmAddition.txtMRatio(3) = frmSolution.txtXe136
   frmAddition.txtMRatio(4) = frmSolution.txtKr82
   frmAddition.txtMRatio(5) = frmSolution.txtKr83
   frmAddition.txtMRatio(6) = frmSolution.txtKr84
   frmAddition.txtMRatio(7) = frmSolution.txtKr85
   frmAddition.txtErrMRatio(0) = frmSolution.txtErrXe130
   frmAddition.txtErrMRatio(1) = frmSolution.txtErrXe131
   frmAddition.txtErrMRatio(2) = frmSolution.txtErrXe132
   frmAddition.txtErrMRatio(3) = frmSolution.txtErrXe136
   frmAddition.txtErrMRatio(4) = frmSolution.txtErrKr82
   frmAddition.txtErrMRatio(5) = frmSolution.txtErrKr83
   frmAddition.txtErrMRatio(6) = frmSolution.txtErrKr84
   frmAddition.txtErrMRatio(7) = frmSolution.txtErrKr85

   j = 0
   For i = 0 To 7
      j = j + 1
      frmAddition.txtCRatio(i) = Format$(Rcalc(iset1, j), "0.0000")
      frmAddition.txtErrCRatio(i) = Format$(eRcalc(iset1, j), "0.0000")
      If j = 2 Then
         j = j + 1
      End If
   Next i

' Output Plutonium Information
   If IsNumeric(frmSolution.txtKr85) And IsNumeric(frmSolution.txtErrKr85) Then
      halflife = 87.7
      Rcalc(iset1, 10) = Rcalc(iset1, 10) * Exp(-Log(2#) * FAge / halflife)
      halflife = 14.4
      Rcalc(iset1, 13) = Rcalc(iset1, 13) * Exp(-Log(2#) * FAge / halflife)
      frmAddition.txtComment2 = "Plutonium compositions are calculated at fuel age (i.e., decay
corrected)"
   Else
      frmAddition.txtComment2 = "Plutonium compositions are calculated at discharge (i.e., not decay
corrected)"
   End If
   TotPu = 0#
   ErrTotPu = 0#
   For i = 0 To 5
      frmAddition.txtPu(i) = Format$(Rcalc(iset1, i + 10), "####0.000")
      frmAddition.txtErrPu(i) = Format$(eRcalc(iset1, i + 10), "####0.000")
      TotPu = TotPu + Rcalc(iset1, i + 10)
      ErrTotPu = ErrTotPu + eRcalc(iset1, i + 10)
   Next i
   frmAddition.txtTotPu = Format$(TotPu, "####0.000")
   frmAddition.txtErrTotPu = Format$(ErrTotPu, "####0.000")

' Output Fission Product and Minor Actinide Information
```

```
    thalf(16) = 2.065
    thalf(17) = 2300000#
    thalf(18) = 30.17
    thalf(19) = 213000#
    thalf(20) = 15700000#
    thalf(21) = 29.1
    thalf(22) = 10000000000#
    thalf(23) = 10000000000#
    thalf(24) = 2140000#
    thalf(25) = 432.7
    thalf(26) = 141#
    thalf(27) = 7370#
    thalf(28) = 162.8 / 365#
    thalf(29) = 29.1
    thalf(30) = 18.1
    thalf(31) = 8500#
    If IsNumeric(frmSolution.txtKr85) And IsNumeric(frmSolution.txtErrKr85) Then
      For i = 16 To 31
        FAge = FAge - (150# / 365#)
        Rcalc(iset1, i) = Rcalc(iset1, i) * Exp(-Log(2#) * FAge / thalf(i))
      Next i
      frmAddition.txtComment1 = "Fission product and minor actinide compositions are at fuel age"
    Else
      frmAddition.txtComment1 = "Fission product and minor actinide compositions are at 150 days
decay"
    End If
    For i = 0 To 7
      frmAddition.txtFP(i) = Format$(Rcalc(iset1, i + 16), "0.000E+00")
      frmAddition.txtErrFP(i) = Format$(eRcalc(iset1, i + 16), "0.000E+00")
      frmAddition.txtMA(i) = Format$(Rcalc(iset1, i + 24), "0.000E+00")
      frmAddition.txtErrMA(i) = Format$(eRcalc(iset1, i + 24), "0.000E+00")
    Next i

End Sub

Private Sub tmrCalculate_Timer()
    frmSolution.Hide
    pval = pval + 10
    If pval > 99 Then
      If jflag = 0 Then
        frmCalculate.Hide
        frmSolution.Hide
        frmIntro.Hide
        frmData.Show
        frmAdvanced.Hide
        frmConfirm.Hide
        Unload frmCalculate
        Exit Sub
      Else
        frmCalculate.Hide
        frmSolution.Show
        frmIntro.Hide
```

```
        frmData.Hide
        frmAdvanced.Hide
        frmConfirm.Hide
        Unload frmCalculate
        Exit Sub
      End If
    End If
    prbCalculation = pval
End Sub
```

frmSolution

```
Option Explicit

Private Sub cmdAddition_Click()
    frmSolution.Hide
    frmAddition.Show
End Sub

Private Sub cmdBack_Click()
    Dim i As Integer
    pflag = 0
    Max = 20
    For i = 1 To Max
      Pvalue(i) = 1
    Next i
    Unload frmSolution
    frmIntro.Hide
    frmData.Show
    frmAdvanced.Hide
    frmSolution.Hide
    frmConfirm.Hide
End Sub

Private Sub cmdDone_Click()
    Dim i As Integer
    pflag = 0
    Max = 20
    For i = 1 To Max
      Pvalue(i) = 1
    Next i
    Unload frmSolution
    Load frmIntro
    Unload frmIntro
    frmIntro.Show
    frmData.Hide
    frmAdvanced.Hide
    frmSolution.Hide
    frmConfirm.Hide
End Sub
```

```
Private Sub cmdPrint_Click()
   Dim FileNum As Integer
   FileNum = FreeFile
   Open txtFilename.Text For Output As FileNum
   Write #FileNum, fraIsos.Caption
   Write #FileNum, lblXe130.Caption, txtXe130.Text
   Write #FileNum, lblXe131.Caption, txtXe131.Text
   Write #FileNum, lblXe132.Caption, txtXe132.Text
   Write #FileNum, lblXe136.Caption, txtXe136.Text
   Write #FileNum, lblKr82.Caption, txtKr82.Text
   Write #FileNum, lblKr83.Caption, txtKr83.Text
   Write #FileNum, lblKr84.Caption, txtKr84.Text
   Write #FileNum, lblKr85.Caption, txtKr85.Text

   Write #FileNum, " "
   Write #FileNum, fraResult.Caption
   Write #FileNum, lblBurnup.Caption, txtBurnup.Text, "+/-", txtError.Text
   Write #FileNum, lblType.Caption, txtType.Text
   Write #FileNum, lblAge.Caption, txtAge.Text
   Write #FileNum, lblHistory.Caption, txtHistory.Text
   Write #FileNum, lblConfidence.Caption, txtConfidence.Text

End Sub
```

frmAddition

```
Private Sub cmdBack_Click()
   frmSolution.Show
   frmAddition.Hide
End Sub
```

APPENDIX D

Measured Xenon and Krypton Data for LWR Fuels

TABLE D.1

Summary of Xenon and Krypton Measurements for PWRs

Reactor Name	Reactor Type	Sample	*Declared or Measured Values* wt% U-235	131/134	132/134	83/86	84/86	85/86	Measured Burnup
Obrigheim	PWR	86	3.1	0.308	0.702				28974
Obrigheim	PWR	87	3.1	0.281	0.725	0.241	0.671		30917
Obrigheim	PWR	88	3.1	0.302	0.746	0.243	0.655		28859
Obrigheim	PWR	89	3.1	0.287	0.704	0.239	0.636		29671
Obrigheim	PWR	90	3.1	0.292	0.720	0.241	0.663		30181
Obrigheim	PWR	91	3.1	0.297	0.714	0.231	0.634		28542
Obrigheim	PWR	92	3.1	0.304	0.710	0.245	0.646		27061
Obrigheim	PWR	93	3.1	0.297	0.737	0.243	0.658		29857
Obrigheim	PWR	94	3.1	0.309	0.706	0.242	0.643		26452
Obrigheim	PWR	95	3.1	0.302	0.715	0.241	0.639		28650
Obrigheim	PWR	d1 p1	3	0.335	0.685				19520
Obrigheim	PWR	e3 p2	3	0.306	0.734	0.233	0.613	0.100	29530
Obrigheim	PWR	e3 p4	3	0.296	0.725	0.231	0.611	0.105	30940
Obrigheim	PWR	g7 p1	3	0.333	0.656	0.255	0.573	0.104	17490
Obrigheim	PWR	g7 p3	3	0.294	0.757	0.229	0.613	0.104	31920
Obrigheim	PWR	g7 p5	3	0.304	0.723	0.245	0.602	0.107	28830
Obrigheim	PWR	m14 p1	3	0.339	0.649	0.257	0.567	0.106	15600
Obrigheim	PWR	m14 p4	3	0.303	0.710	0.241	0.594	0.105	27460
Obrigheim	PWR	g14 p31	3	0.275	0.761	0.212	0.631	0.104	37490
Borssele	PWR	a8	3.1	0.295	0.729	0.221	0.602	0.109	31000
Borssele	PWR	o6	3.1	0.290	0.733	0.231	0.608	0.106	31800
Borssele	PWR	g15	3.1	0.288	0.730	0.218	0.614	0.107	31600
Borssele	PWR	i15	3.1	0.287	0.724	0.226	0.601	0.107	31500
Borssele	PWR	a10	3.1	0.287	0.729	0.224	0.599	0.107	31000

TABLE D.2

Summary of Xenon and Krypton Measurements for BWRs

Reactor Name	Reactor Type	Sample	*Declared or Measured Values* wt% U-235	131/134	132/134	83/86	84/86	85/86	Measured Burnup
Garigliano	BWR	a1	1.6	0.354	0.668	0.263	0.576	0.118	10590
Garigliano	BWR	a9	1.6	0.339	0.682	0.265	0.590	0.114	14040
Garigliano	BWR	b1	1.6	0.351	0.665	0.267	0.592	0.116	9800
Garigliano	BWR	j1	1.6	0.342	0.682	0.267	0.596	0.116	12830
Garigliano	BWR	j9	1.6	0.337	0.686	0.268	0.597	0.118	14480
Garigliano	BWR	a3	2.1	0.349	0.650	0.263	0.567	0.115	10510
Garigliano	BWR	b2	2.1	0.351	0.655	0.268	0.570	0.117	10280
Garigliano	BWR	b8	2.1	0.343	0.661	0.266	0.571	0.114	12150
Garigliano	BWR	c1	2.1	0.349	0.653	0.264	0.567	0.111	10660
Garigliano	BWR	c3	2.1	0.349	0.652	0.271	0.569	0.119	9140
Garigliano	BWR	d2	2.1	0.349	0.652	0.270	0.568	0.110	9440
Garigliano	BWR	d4	2.1	0.349	0.679	0.270	0.565	0.118	8850
Garigliano	BWR	e1	2.1	0.348	0.657	0.265	0.573	0.112	10800
Garigliano	BWR	e5	2.1	0.346	0.655	0.263	0.568	0.115	8930
Garigliano	BWR	g7	2.1	0.349	0.659	0.270	0.570	0.118	10540
Garigliano	BWR	h2	2.1	0.342	0.661	0.260	0.568	0.114	11920
Garigliano	BWR	h8	2.1	0.347	0.667	0.262	0.586	0.119	12700
Gundremmingen	BWR	b3	2.53	0.303	0.698	0.245	0.588		21240
Gundremmingen	BWR	c5	2.53	0.307	0.714	0.249	0.594		22970
Gundremmingen	BWR	e5	2.53	0.294	0.711	0.258	0.581		25190
Gundremmingen	BWR	b3	2.53	0.334	0.675	0.256	0.571		14390
Gundremmingen	BWR	c5	2.53	0.327	0.666	0.254	0.573		15840
Gundremmingen	BWR	e5	2.53	0.328	0.675	0.249	0.581		17490

VITA

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