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NUCLEAR FORENSICS TECHNIQUE FOR ATTRIBUTING MATERIAL USED IN A RADIOLOGICAL DISPERSAL DEVICE EVENT

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ABSTRACT

If a radiological dispersal device (RDD) is detonated in the U.S. or near U.S. interests overseas, it will be crucial that the actors involved in the event can be identified quickly. Law enforcement officials will need information concerning the material used in the device, specifically what type of material it was and from where it originated. This information will then be used to help identify the specific individuals who manufactured the device and perpetrated the event. Texas A&M University and Los Alamos National Laboratory are collaborating on the development of a technique for identifying the material used in a radiological dispersal device. This methodology is currently focused on radiological dispersal devices that make use of spent nuclear fuel as the source material. The methodology developed makes use of both a forward model and an inverse model to identify specific spent fuel characteristics using isotopic composition of RDD debris. The forward model is based on sophisticated reactor physics calculations for the prediction of spent fuel isotopic compositions as a function of fuel type (e.g., PWR, BWR, CANDU, RBMK, etc.), fuel burnup (in MWd/MTHM), fuel age (in years since permanent discharge from the reactor), and operating characteristics (e.g., operating power level, time at power, etc.). These reactor physics calculations are benchmarked to measured data to establish their accuracy in predicting isotopic compositions. The inverse model makes use of a Bayesian inverse method to identify the specific spent fuel assembly (or assemblies) used based on measurements of actinide and fission product isotopic ratios in the RDD debris. A description of both the forward and inverse models, accuracies of the technique, and the results to date are given.

INTRODUCTION

The events of September 11, 2001 clearly show the willingness of terrorists to use conventional means for inflicting great casualties. Nuclear terrorism is one of those possible means. While it is unlikely that terrorist groups would have the capability to fabricate a nuclear weapon, these groups would likely have the capability to produce a Radiological Dispersal Device (RDD, or the so-called "dirty bomb"). An RDD is "any device, other than a nuclear explosive device, specifically designed to employ radioactive material by disseminating it to cause destruction, damage, or injury by means of the radiation produced by the decay of such material" [1].

While it is unlikely that the detonation of an RDD would inflict a large number of casualties, its terror effect would be immense and it could be used to deny use of an area. The threat of a terrorist using (RDD) inside the U.S. or against U.S. interests overseas is greater than ever. This is due both to the increased sophistication of terrorist organizations and to the large amount of nuclear and radiological material at use or in storage throughout the world. All of the following are possible sources of material for an RDD:

1. Large radioactive sources for industrial radiography (mostly radium, ^{192}Ir, or ^{60}Co)
2. Large neutron sources (PuBe, AmBe, AmLi, or ^{252}Cf)
3. Source for food irradiation (^{137}Cs or ^{60}Co)
4. Medical isotopes
5. Radioisotope thermal-electric generators (^{90}Sr or ^{238}Pu)
6. Well-logging sources (mostly AmBe or PuBe, may also have ^{137}Cs)
7. Spent nuclear fuel
8. Reprocessing facility waste products

Generally these materials can be divided into two categories: (a) *large radioactive sources* and (b) *spent fuel sources*. Numbers 1-6 above are all examples of *large radioactive sources*. These materials are fairly well characterized and generally consist of only one major radioisotope and potentially several impurities or trace isotopes. These sources vary from highly radioactive to low-levels of radioactivity. The security involved in guarding these sources typically ranges for low-to-medium level.

Numbers 7 and 8 above are examples of *spent fuel sources*. These sources of material are poorly characterized in their current state though they were well characterized prior to irradiation in a nuclear reactor. These sources are highly-radioactive and even a single fuel assembly would kill anyone exposed directly to its radiation. The level of security in place for these sources is generally high.

If someone acquired one of these materials, fabricated an RDD, and detonated it, it would be crucial that the perpetrator of this act be determined. To identify the actors involved in this event, forensic evidence will be used to build a case against an individual or group. One principal piece of forensic evidence is the origin of the material used in the device (i.e., the name and location of the facility from which the material was acquired). This origin will be determined using the attributes of the material at the time of detonation and matching those attributes to expected attributes of materials stored worldwide. While various attributes will be used, we will limit our discussion here to using isotopic compositions of the residue.

In the case of *large radioactive sources*, establishing the material origin involves determining the isotopic composition of the radioactive material used and any trace or impurity isotopes in the material. The type of material used (e.g., ^{60}Co) will allow for determination of a number of possible suspect origins. Then the trace isotopes will be used to determine the most likely specific origin of the material. While the data necessary to identify the origin of the material is immense, the mathematical means for finding a most likely origin is well developed. Due to this characteristic, this paper will focus on the second type of material: "spent fuel sources".

For spent fuel sources, the isotopic composition of the material at detonation is generally unknown. Thus a measurement of the isotopic composition of the debris will not be directly compared to a database of isotopic materials to identify the source of the RDD. Instead these isotopic compositions will be used to determine the characteristics of the reactor that was used to produce the material. Specifically the reactor type (e.g., VVER-440, RBMK-1500, BN-350, etc.), fuel burnup, fuel age, initial fuel enrichment, and operational history will be determined. These characteristics will then be matched to known reactor systems to determine the most likely system that could have produced this material. It should also be noted that distinguishing between separated (i.e., reprocessed material) and unseparated material is simply a function of identifying the lack of expected isotopes (e.g., the lack of any fission product nuclides).

NUCLEAR FORENSICS PROJECT OVERVIEW

The determination of the attributes of the spent fuel material involves three fundamental components:

1. A forward model for the determination of spent fuel isotopic composition given the design characteristics and operating history of a known reactor facility. This model performs isotopic inventory calculations forward with time. The forward model is based on well-developed reactor physics techniques for calculating spent fuel compositions.
2. A database of known reactor facilities that includes numerous facility characteristics including fuel designs, operational history, and refuelling schedules. This database (named Sentry) is maintained by Los Alamos National Laboratory (LANL) and contains an enormous amount of information concerning nuclear facilities worldwide.
3. An inverse model that can use measured isotopic signatures from RDD residue to predict the most likely characteristics of the facility used to produce those signatures.

A general overview of the forensics methodology used in this project is given in Figure 1. Because of the large amount of material available and the lower security of the material, Russian reactors are being considered first in this project; however, other reactors will be added in the future.

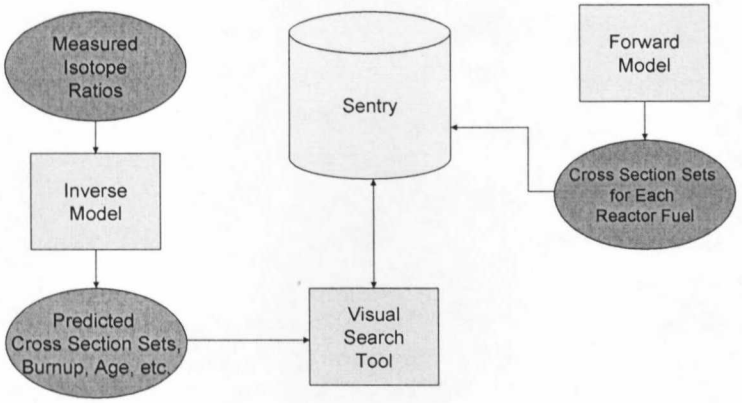

Figure 1. Forensics project overview.

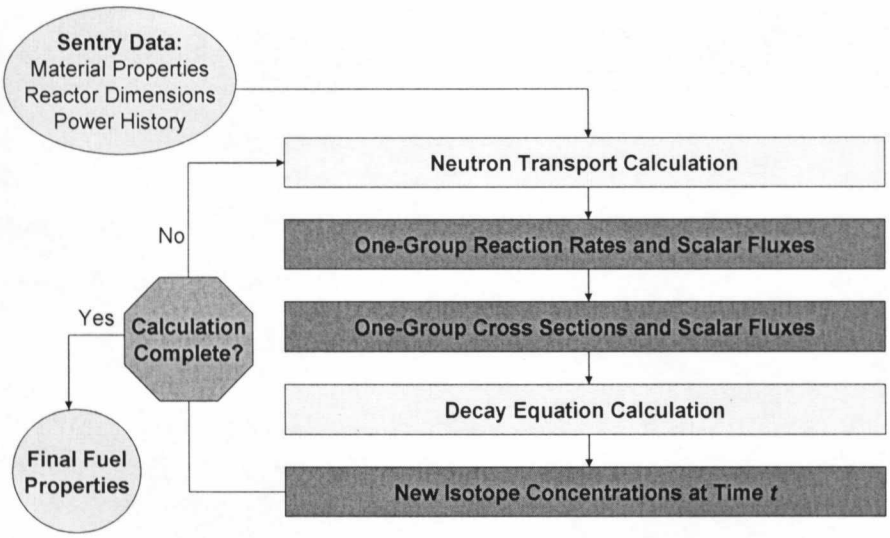

Figure 2. Forward model dataflow.

FORWARD MODEL

The forward model is used to calculate the isotopic composition of spent fuel given the initial fuel dimensions, initial fuel material compositions, the fuel power history, and the fuel decay time (i.e., the time since permanent discharge from the reactor). A schematic of the forward model used is shown in Figure 2. This model is commonly used in present day reactor physics calculations and can be benchmarked to provide accurate results. Due to the diverse types of reactors considered in this study, the Monteburns code system was chosen to implement this forward model [2]. Monteburns uses MCNP-5 to calculate the one group cross sections and scalar fluxes for the fuel and ORIGEN2 to solve the burnup and decay equations for the fuel isotopics [3, 4]. Due to the usage of MCNP-5, Monteburns can simulate essentially any known fuel design.

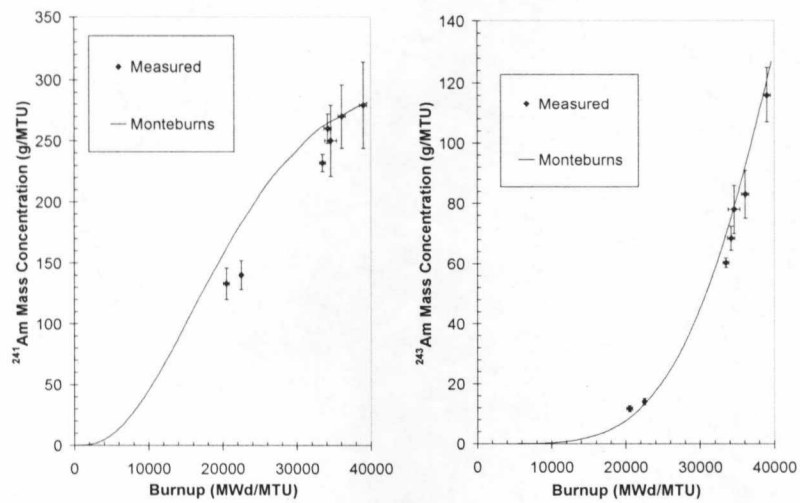

Figure 3. Comparisons between selected measured and Monteburns calculated spent fuel isotopic compositions for a VVER-440 spent fuel.

Forward models have been completed for all of the following reactor types: VVER-440, VVER-1000, RBMK-1500, BN-350, BN-600, and IRT reactors. Also, models for several western-style PWR and BWR reactors have also been completed for testing purposes. The uncertainty in isotopic compositions predicted by Monteburns is being developed by benchmarking the Monteburns code system, and its particular implementation for this project, to measured data from the literature. Figures 3 and 4 show some of the data for this benchmarking. As can be seen good agreement is found between the measured and calculated isotopics. Generally, the forward model has the following accuracies for isotope ratio predictions: $^{235}U/^{238}U$ is ±2.0%, $^{239}Pu/^{238}U$ is ±2.5%, $^{240}Pu/^{239}Pu$ is ±3.2%, $^{237}Np/^{235}U$ is ±5%, $^{241}Am/^{238}U$ is ±10%, $^{243}Am/^{238}U$ is ±14%, $^{148}Nd/^{238}U$ is ±2.3%, $^{237}Cs/^{238}U$ is ±2.4%, and $^{241}Pu/^{238}U$ is ±3.4%.

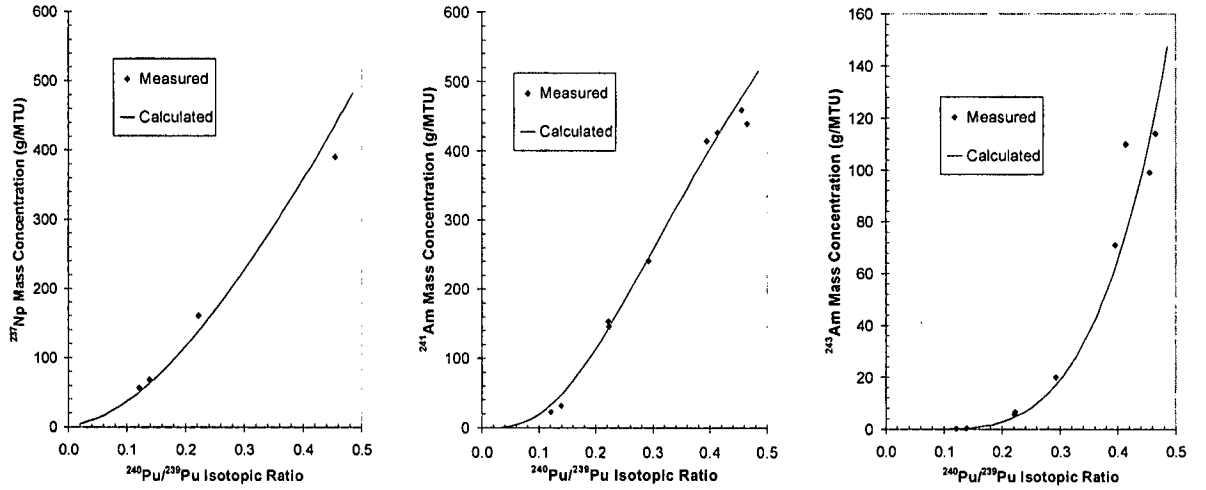

Figure 4. Comparisons between selected measured and Monteburns calculated spent fuel isotopic compositions for a Westinghouse-style PWR.

INVERSE MODEL

The inverse model used in this project is based on analytical inversions of the burnup equations. We use specific monitors for each attribute of interest. To increase the accuracy of the analytical method all monitors were normalized to the ^{238}U concentration. This also will allow for accurate mass spectrometric measurements. The following specific monitors are used:

1. Burnup monitors. These are stable or long-lived isotopes that are invariantly produced under most conditions (i.e., a certain percentage of this monitor is produced per fission that occurs in the fuel). $^{148}Nd/^{238}U$ is an excellent example of this monitor.
2. Reactor type monitors. This type of monitor varies significantly based on the neutron spectrum in the fuel and the fission isotopes in the fuel. $^{104}Ru/^{238}U$ is an an example of this monitor.
3. Fuel age. This type of monitor is produced directly from fission and has a short enough half-life that some portion of it decays between discharge from the core and use as an RDD. $^{241}Pu/^{238}U$ and $^{154}Eu/^{238}U$ are examples of this type of monitor.

4. Fuel enrichment. This monitor is used to determine the initial fuel enrichment and consists of a number of actinide isotopes normalized to ^{238}U. $^{235}U/^{238}U$ and $^{239}Pu/^{238}U$ are examples of this type of monitor.
5. Fuel specific power level and power history. These monitors have characteristics (like short half-lives) that allow them to be used to predict the specific power of the fuel and the fuels power history. $^{134}Cs/^{238}U$ is an example of this type of monitor.

Each of these monitors is used in a hierarchical fashion as shown in Figure 5. This implies that information from one attribute monitor is fed to the subsequent attribute monitor. The uncertainties associated with each calculate are propagated and the most likely set of attributes are predicted. A Bayesian methodology is then used to predict the most likely reactor from the database to be the source of the material.

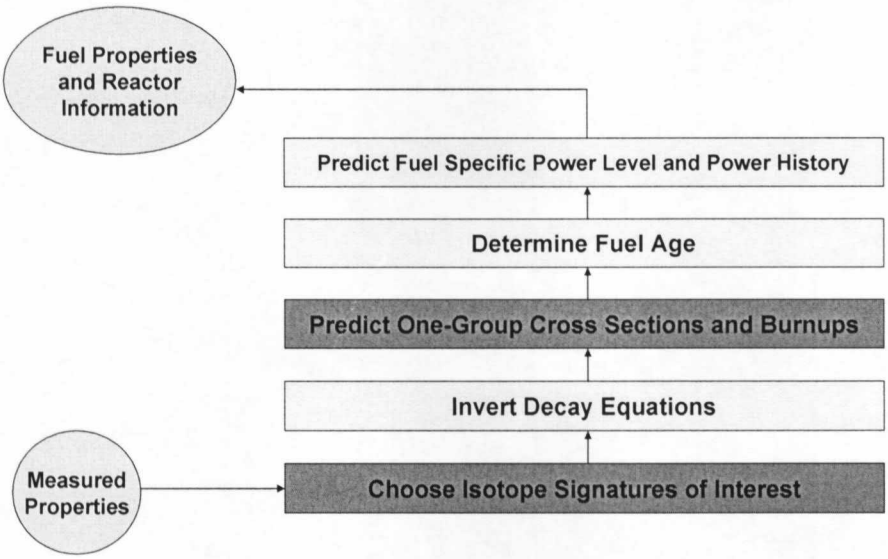

Figure 5. Inverse model dataflow.

PRELIMINARY RESULTS

To test the methodology developed, a simulated experiment was conducted in which a U.S. pressurized water reactor (PWR) fuel with an initial fuel enrichment of 3.20 w/o was simulated to a burnup of 33,000 MWd/MTU. The fuel was irradiated in a standard three-cycle irradiation with 60 day shutdowns between each cycle. Following discharge the fuel was allowed to decay for 16.0 years. All simulations were performed using ORIGEN2. The isotopic composition after decay was assumed to be what was present at the time of detonation in an RDD. The following isotopic ratios were used: $^{235}U/^{238}U$, $^{236}U/^{238}U$, $^{239}Pu/^{238}U$, $^{240}Pu/^{238}U$, $^{241}Pu/^{238}U$, $^{237}Np/^{238}U$, $^{148}Nd/^{238}U$, $^{101}Ru/^{238}U$, $^{104}Ru/^{238}U$, and $^{243}Am/^{238}U$. Measurement uncertainty was added to these calculated values by adding a 0.5% Gaussian error to each isotopic ratio. The predicted and true attributes for the fuel are shown in Table I. Generally good agreement was found except that the system had difficulty distinguishing between PWR and BWR fuel types. This is due to the fact that PWR and BWR neutron spectra and isotopic compositions are generally very similar. More effort is needed to resolve this problem by including additional reactor type monitors.

TABLE I
Predicted and True Attributes for Simulated Experiment

Attribute	Predicted	True	%Error
Fuel burnup	33609	33000	1.85%
Fuel type	PWR or BWR	PWR	n/a
Fuel age	16.3	16.0	1.88%
Fuel enrichment	3.16	3.20	1.25%

The results in Table I are promising. The estimation of 0.5% measurement error is reasonable for some isotopes; however, a more realistic experiment would have been to include different errors (both random and systematic on each isotopic ratio). It is expected that the percent errors for the predicted values from the true values will increase under this scenario. Also, it should be noted that no attempt was made to match this attributed fuel to a reactor in the database.

CONCLUSIONS AND FUTURE WORK

A description of a nuclear forensics methodology that can be used for the attribution of material used in an RDD was given. This method was developed through a collaborative project between Texas A&M University and LANL. The methodology described focused on RDDs that used spent nuclear fuel as their source material; however, the capability of attributing material for RDDs with other material is also under development. Descriptions of both the forward and inverse models were given as well as some preliminary results. The preliminary results using 10 different isotopic ratios are promising and suggest that an RDD composed of spent fuel source material could be properly attributed. Future effort will be spent identifying additional reactor type monitors and in establishing rigorous uncertainty estimates.

It should be noted that though the discussion above was focused on analysis of material post-detonation, all of the above could be used in a pre-detonation scenario as well (i.e., if the RDD was discovered and rendered harmless before it was detonated).

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