# REACTOR PHYSICS MODELING OF SPENT NUCLEAR RESEARCH REACTOR FUEL FOR SNM ATTRIBUTION AND NUCLEAR FORENSICS

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## ABSTRACT

Nuclear research reactors are the least safeguarded type of reactor; in some cases this may be attributed to low risk and in most cases it is due to difficulty from dynamic operation. Research reactors vary greatly in size, fuel type, enrichment, power and burnup providing a significant challenge to any standardized safeguard system. If a whole fuel assembly was interdicted, based on geometry and other traditional forensics work, one could identify the material's origin fairly accurately. If the material has been dispersed or reprocessed, in-depth reactor physics models may be used to help with the identification. Should there be a need to attribute research reactor fuel material, the Savannah River National Laboratory would perform radiochemical analysis of samples of the material as well as other non-destructive measurements. In depth reactor physics modeling would then be performed to compare to these measured results in an attempt to associate the measured results with various reactor parameters.

Several reactor physics codes are being used and considered for this purpose, including: MONTEBURNS/ORIGEN/MCNP5, CINDER/MCNPX and WIMS. In attempt to identify reactor characteristics, such as time since shutdown, burnup, or power, various isotopes are used. Complexities arise when the inherent assumptions embedded in different reactor physics codes handle the isotopes differently and may quantify them to different levels of accuracy. A technical approach to modeling spent research reactor fuel begins at the assembly level upon acquiring detailed information of the reactor to be modeled. A single assembly is run using periodic boundary conditions to simulate an infinite lattice which may be repeatedly burned to produce input fuel isotopic vectors of various burnups for a core level model. A core level model will then be constructed using the assembly level results as inputs for the specific fuel shuffling pattern in an attempt to establish an equilibrium cycle. The core level results may then be compared to the radiochemistry results from the dissolved fuel samples and a decision whether further more in-depth modeling should be performed.

The SRNL is in the process of analyzing multiple research reactor fuels to determine the best means to provide forensic data for attribution and assess codes and modeling methods for attribution. As several fuel samples are analyzed, this work will allow improved SNM forensics of spent research reactor fuel. This will enable the establishment of a research reactor fuel database of SNM materials, and allow an attempt of an inverse analysis if research reactor material is diverted and seized.

## INTRODUCTION

Nuclear forensics is the technical means by which nuclear material is characterized and interpreted. These may suggest or exclude possible sources of origin when attempting to attribute a nuclear device, designs, or material to a source of origin (1).

Pre-detonation material analysis and attribution of spent research reactor fuel provides a challenging nuclear forensics problem. The tremendous variation between research reactor designs as well as dynamic operation lead to the existence of spent fuel containing a great variety of burnups, enrichments, power history, shape and material form. Research reactors are also the least safeguarded type of reactor. This is due to a perception of low risk and/or difficulty in applying safeguards on reactors with such dynamic operations. Combined with locations that tend to encourage openness, university campuses for example, make research reactors a potential target. A common type of research reactor is the pool type, consisting of a reactor immersed in an open cooling pool of water, which usually operate around or below approximately 10 $MW_{th}$. This type of reactor is often composed of either HEU or LEU in an aluminum alloy and can use many types of fuel including: plates, rods, and tubes (2). Other types of research reactors have power levels greater than pool type reactors and require more powerful heat extraction systems. Both of these types of research reactors may possess HEU fuel enriched to greater than 90% $^{235}U$. The International Nuclear Fuel Cycle Evaluation report in 1980 shows more than 150 research reactors in more than 35 different countries operated using HEU fuels at power levels between 10 $kW_{th}$ and 250 $MW_{th}$ (3). Programs have been in progress for many years to convert most of the HEU reactors to LEU fuels, for the purpose of reducing proliferation risks.

Two classifications of weapons of mass destruction are considered: a fission type nuclear weapon and a radiation dispersal device. Both cases are relevant as the broad variety of spent research reactor fuel includes highly enriched uranium which could be used for a nuclear yield device and enough activity for an effective radiation dispersal device. Most research reactors operate at low power levels, less than 10 $MW_{th}$, which is not optimal for producing plutonium (3). In the case of a nuclear weapon, the spent fuel and fresh fuel may be classified as direct use material depending on $^{235}U$ enrichment, but possess different safeguards timeliness goals (4). In the case of a radiation dispersal device, the material may directly be used and simply placed alongside high explosives to disperse activity.

If material of research reactor origin was diverted and seized, using the material characteristics with minimal uncertainties, an inverse analysis may be applied to attempt to determine the material's origin. If perfect measurements of the materials characteristics were present, the inverse analysis may produce a well-posed problem. Then a reconstruction of the reactor parameters to determine the materials origin may be simple, assuming a unique solution exists. As uncertainties propagate through any inverse problem, it is expected that the results worsen and reconstruction ability will diminish.

## RESEARCH REACTOR SIGNATURE DATABASE

An effort is in progress to produce a database of research reactor signatures that may be classified into two categories the first including measured or recorded signatures and the second being modeled signatures. Measured or recorded signatures may be created from measured data, reactor operators logs and manuals, and other documented information. Modeled signatures are necessary

for reactors where this data is not possible to obtain, but instead it is digitally created using reactor physics modeling.

To establish a database of reactor signatures and spent fuel characteristics by modeling with reactor physics codes, a series of limiting case reactors will be analyzed in depth. These reactors will go through a multistep analysis process involving reactor physics modeling and radiochemistry processes. The L-basin at Savannah River Site possesses domestic spent research reactor fuel of many types. Particular fuel assemblies will be chosen based on limiting criteria with respect to burnup, $^{235}U$ content, and activity.

Upon selection, a sample of the spent fuel will be collected and radiochemistry analysis performed. The fuel will simultaneously be modeled using reactor physics depletion codes. The main two reactor physics software packages used in this analysis are MONTEBURNS/ORIGEN/MCNP5 and MCNPX/CINDER. MONTEBURNS is a Perl script tool that links the Monte Carlo neutral particle MCNP with the radioactive decay and burnup code ORIGIN 2.2. The software automates the creation of input files to be passed back and forth between the two codes depending on user specified burn information. It requires multiple input files including geometry and material inputs in MCNP and a MONTEBURNS format input consisting of depletion and material feed parameters (5). CINDER is a routine package bundled within the latest version of MCNPX which performs all of the similar computations in a standalone package. This package only requires a similar MCNP geometry and material input but also requires the addition of a BURN card to specify the depletion parameters.

The level of detail in the model required to predict and verify the radiochemistry results will be determined from several different methods. Initially, a simple infinite lattice of an assembly model will be performed. If this model does not agree with the radiochemistry results, then a more detailed model will be explored, such as full core model. The output isotopic vectors from the assembly level model would be useful as the input for assemblies in a core model at different burnup steps. Upon completion of the core, various external core features may be added such as a core housing, surrounding water, and concrete. This full core model may be run through multiple burnup steps while rotating and updating the fuel isotopic composition each step until an equilibrium cycle is obtained. Once the level of detail required to produce accurate reactor characteristics is determined, the process may continue with modeling alone and the establishment of a more complete modeled signature database may proceed.

## INVERSE PROBLEM METHODS

In the case that material was acquired and attributions methods are needed, there are several ways to attempt an identification of the material's origin. There are two main categories of methods appropriate in this case: discrete and continuous numerical optimization methods.

There are two ways to pose this as a numerical optimization problem; a discrete optimization approach and a continuous optimization approach. Both of these methods involve the reconstruction of a research reactor signature set that will be used to determine the material's origin. These inverse methods would require an input of material characteristics from a sample including but are not limited to: isotopic information, material form, material shape and size characteristics. The reconstructed signature set would include characteristics such as fuel burnup, final and initial fuel

$^{235}U$ enrichment, reactor power, time elapsed since fuel discharge, etc. which would then be used to attempt to determine the origin of the material.

In discrete optimization, a predefined solution space is required prior to the analysis which is contained in a finite set (6). In this case, a set database of all research reactors and research reactor material characteristics must be known prior to implementation. During the inverse analysis, the current iterate solution would limit the solution to the source of origin which is closest to the optimality conditions. This method may provide a quality estimate of material origin, but is limited by the database information and sample measurement uncertainty. If the solution is far from any dataset entry, expert judgment must be applied.

A particular type of discrete optimization method is a database search algorithm, which may be used to pair a sample with a database entry if a database entry exists. There are many variations of database search algorithms which may be applicable depending on what the database information consists of. If the database contains complete spent fuel characteristics and a spent fuel sample is obtained, a simple algorithm may just locate the closest match. This method could search both types of signature databases to obtain results.

Another way to approach this problem is using continuous optimization. In continuous optimization, no complete predetermined solution space of the material characteristics is required where the solution that minimizes the optimality conditions is chosen (6). Many solutions may exist that satisfy these optimality conditions and constraints may be applied to limit solutions to realistic cases. Using this category of optimization, specific reactor parameters may be reconstructed numerically, then if no actual reactor parameter sets or multiple reactor parameter sets meet the reconstructed solutions, expert judgment may be implemented.

As measurement uncertainties may propagate, a conclusion of a single source of origin may not arise. If using a database search algorithm, there may be multiple solutions in the search result. If the problem is posed as a numerical optimization problem, there may not be a unique solution but in either case the methods may allow a significant quantity of solutions to be excluded. This will narrow down possible sources of origin if a sample is obtained. If a sample did not contain all of the desired characteristics, for example had been separated or blended, a much more difficult problem arises and conclusions may not be able to be made even if measurement uncertainty approaches zero.

## PRELIMINARY RESULTS

The first reactor assembly to be analyzed is the Oak Ridge Reactor (ORR) which consisted of the following properties shown in Table 1. This reactor operated at a full power of 30 $MW_{th}$ until final shutdown on March 26, 1987 and consists of materials test reactor (MTR) plate fuel elements.

Table 1. Oak Ridge Reactor spent fuel assembly characteristics.

| **Assumption/Premise** | **Assumed Value** |
|---|---|
| Assembly Time at Power [d] | 108.9 |
| Assembly Burnup [MWd] | 121 |
| Assembly Power [$MW_{th}$] | 1.111 |
| Fuel Meat Material | $U_3O_8$ & Al |
| Cladding Material | Al |
| Moderator | $H_2O$ |
| Fuel Meat Thickness [cm] | 0.0508 |
| Plate Thickness [cm] | 0.1638 (2 outer) & 0.1275 (17 inner) |
| Active Fuel Length [cm] | 59.8043 (2 outer) & 59.7839 (17 inner) |
| Reactor Cycle Specifics | 239.6 day cycle with 21.78 days at power and 217.82 cooling days |
| Assembly Decay Time [d] | 8799.82 |
| Number of Plates per Assembly | 19 |
| Initial $^{235}U$ [g] | 285 |
| Initial $U_{tot}$ [g] | 306 |
| Final $^{235}U$ [g] | 132 |
| Final $U_{tot}$ [g] | ~177 |
| Total Decay heat [W] | 2.562 |
| Total Activity [Ci] | 882 |

This fuel was modeled on an assembly level and configured in an infinite lattice using periodic boundary conditions in MNCP. To achieve an infinite lattice configuration of an assembly of MTR type with curved fuel plates, one half of the fuel assembly was translated, the distance of the assembly to assembly pitch, to one side to form a rectangular parallelepiped cell. A visualization of this configuration can be seen in Figure 1.

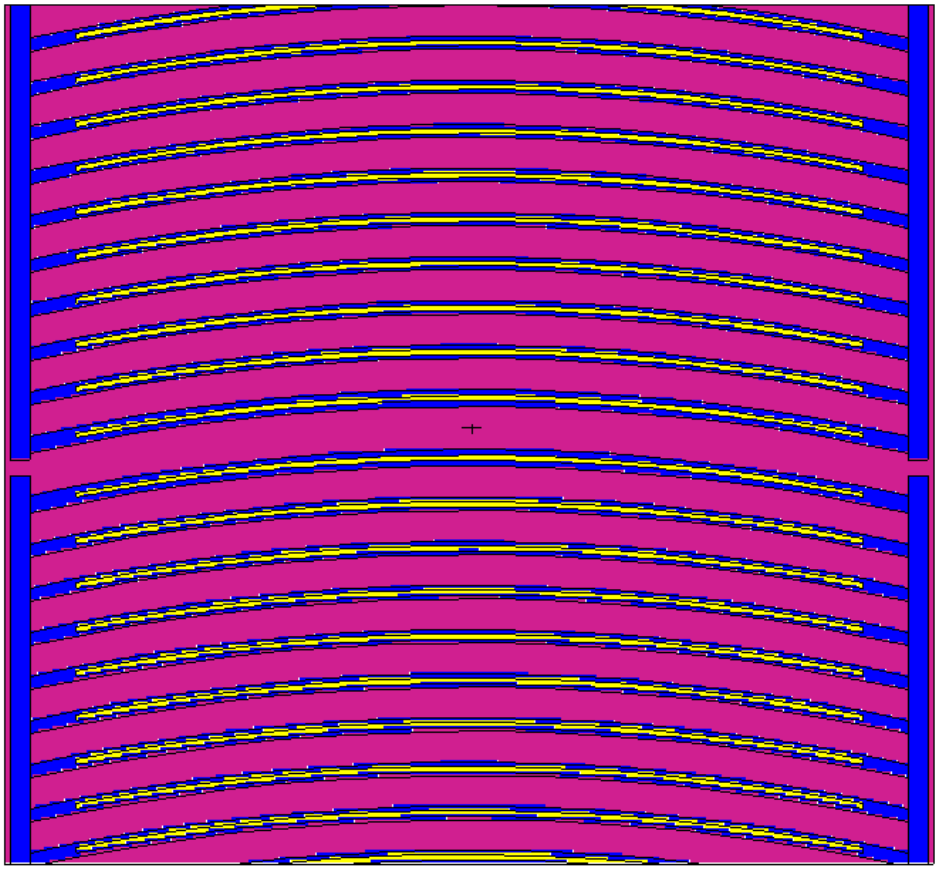

Figure 1. ORR fuel assembly in an infinite lattice configuration.

In Figure 1, the magenta cells represent water in coolant channels and surrounding water, the blue cells represent aluminum cladding and sideplates, and the yellow cells represent the $U_3O_8$ blended with aluminum.

Using MONTEBURNS, a ORIGEN 2.2/MCNP5 depletion routine, this infinite lattice model was burned at the specified power for the entire full power time length of 108.9 days. The effective multiplication factor ($k_{eff}$) including Monte Carlo computation uncertainties, flux, and $^{235}U$ content of this model is shown as a function of burnup in Table 2.

Table 2. MONTEBURNS depletion results as a function of time at full power.

| Timestep [d] | $k_{eff}$ | Flux [n/cm$^2$-s] | $^{235}$U Content [g] (end of step) |
|---|---|---|---|
| 0.00 | 1.527 ± 0.004 | 1.22E+15 | 285.00 |
| 0.00 – 21.78 | 1.431 ± 0.004 | 1.30E+15 | 253.40 |
| 21.78 – 239.6 | 1.431 ± 0.004 | 1.30E+15 | 253.40 |
| 239.6 – 261.38 | 1.431 ± 0.004 | 1.27E+15 | 223.20 |
| 261.38 – 479.2 | 1.467 ± 0.004 | 1.27E+15 | 223.20 |
| 479.2 – 500.98 | 1.381 ± 0.005 | 1.32E+15 | 195.00 |
| 500.98 – 718.8 | 1.439 ± 0.004 | 1.32E+15 | 195.00 |
| 718.8 – 740.58 | 1.354 ± 0.004 | 1.38E+15 | 165.90 |
| 740.58 – 958.4 | 1.406 ± 0.004 | 1.38E+15 | 165.90 |
| 958.4 – 980.18 | 1.310 ± 0.004 | 1.46E+15 | 137.90 |
| 980.18 – 9780 | 1.360 ± 0.004 | 1.46E+15 | 137.90 |

The same MCNP model was run using the MCNPX/CINDER burnup package to produce uranium data at the end of life of the fuel. This package provides a final $^{235}$U content of 134 g which produces a percent difference of 2.8%. It is safe to say that both routines are depleting the $^{235}$U properly as they produce very similar results with a different number of burn steps while holding the time at full power constant.

A full core model of this reactor is still in development while the material inputs are being created by burning the assembly level model repeatedly for each burnup step. Other comparison isotopes will also be benchmarked between MONTEBURNS and the MCNPX/CINDER packages.

The dissolution process of the fuel samples has been scheduled to begin in June 2010.

## CONCLUSIONS

The current state of nuclear security and safeguards instituted at research reactors provides a less rigorously secured pathway toward acquiring nuclear material. While this may be a simpler pathway toward nuclear material, the quantity of material located at any given facility will be often significantly smaller than commercial or weapon facilities. However, in the case of research reactors using HEU fuel, the reactor potentially is a more appealing target than commercial facilities. The vast variety of research reactor types provides spent fuel material useable in many types of WMDs primarily a nuclear weapon and a radiation dispersal device.

To establish modeled signatures for a research reactor database via reactor physics modeling, several specific limiting cases will be involved in a two part process. This process will involve spent fuel sample dissolution and reactor physics modeling to predict and verify these radiochemistry results. This will establish the minimum model detail required to model the fuel to produce spent fuel characteristics with modeling alone.

If a database of research reactor parameters and spent fuel characteristics were established, either a database search algorithm or a numerical optimization method may be used to suggest or exclude

possible sources of origin for an obtained sample of research reactor material. The more complete database acquired, the greater knowledge of a materials origin may be obtained and provide the most benefit to nonproliferation efforts (1).

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