

## **Estimation Procedures and Error Analysis for Inferring the Total Plutonium Produced by a Graphite-Moderated Reactor**

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### **Problem Statement**

This paper describes (1) statistical estimation procedures used in GIRM (Graphite Isotopic Ratio Method), and (2) a methodology for evaluating GIRM's uncertainty.

GIRM estimates the total plutonium (Pu) produced by a graphite-moderated reactor. GIRM accomplishes this by measuring the isotopic ratios of trace elements present in the graphite moderator and using a reactor physics code to relate those isotopic ratios to local Pu production.

More specifically, in the GIRM methodology, graphite samples are taken along the fuel channels in the reactor of interest, and isotopic ratios in the samples are measured with mass spectrography. These isotopic ratios are then converted into local plutonium production rates (i.e. grams of Pu produced per cm of fuel rod) using a reactor physics code (WIMS). Finally, these rates are then integrated over the reactor core to produce a total plutonium estimate.

### **Background**

The basic scheme was proposed ten years ago, and has been subjected to several feasibility studies and experimental tests. Two previous error analyses of this methodology have been conducted. The earliest analysis, (Talbert 95) evaluated uncertainties associated with a "generic" reactor. The other (Reid 97) evaluated actual measurements taken from a British reactor, Trawsfynydd. Our motive is to update the previous work because the GIRM methodology has changed substantially in the ensuing years. In particular:

There is interest in reactors with short operating lives (short-lived reactors), which use a different set of indicator elements for Pu estimation than long-lived reactors.

The GIRM methodology now uses improved sample preparation methods and different mass-spectrographic methods (SIMS plus TIMS) for isotopic analysis. This has required a corresponding modification in the statistical estimation methodology.

Additional sources of error/bias have been identified as a possible concern and need to be accounted for in the error analysis. These sources of error include sample contamination, sample positioning error, calibration error, errors in the 3D fluence model, and errors in the WIMS isotopic ratio calculations.

Another motive for producing a new error analysis of GIRM is to produce results that are logically simpler and more self-contained. The procedures to be described here are illustrated by applying them to a "generic" reactor, which is described in detail in Appendix A.

### Summary of Results

We provide a detailed statistical description of the GIRM estimation algorithms used to produce a total Pu estimate from isotopic ratios. We include some discussion of the assumptions these algorithms rest upon, as well as their statistical properties.

The estimation procedure consists of three separate algorithms. The first algorithm transforms the TIMS U/Pu isotopic ratios into a set of local Pu-fluence estimates using the WIMS reactor computer code. The second transforms the SIMS Boron measurements into a second set of local Pu-fluence estimates. The third algorithm combines the two sets of local fluence estimates to produce a best estimate of total Pu production. This requires an estimate of the 3-D fluence field. Fluence (gm/cm) is the total Pu produced per unit length at a location  $x$  in a fuel channel.

In order to evaluate the uncertainties of the proposed estimation procedure, a Monte Carlo program has been constructed. The strategy is to use the Monte Carlo program to simulate realistic reactor parameters and measurement data which can then be run through the estimation algorithm to allow an assessment of the accuracy of Pu production estimates. We describe the simulation steps that define the Monte Carlo program and also describe the probabilistic error models of the measurement process.

The estimation algorithms produce a standard error as well as the total Pu estimate, using propagation of error methods. The propagation of error methods used here determine the error in an estimate by linearizing the estimation formula and then "propagating" the variances associated with the input variables to the estimate. Such methods are approximate calculations that become worse as the estimation formula becomes more non-linear, and the input variables non-Gaussian. Also the present "propagation of error" variance does not account for all correlations present in the data (such as correlations due to calibration or WIMS code biases), so the Monte Carlo calculation can be used to see how severe this effect may be. The performance of the present propagation of error formulas could probably be improved upon, and the benchmark one would use to measure this improvement would be the Monte Carlo code results.

The current version of GIRM contains the following alterations from earlier versions:

- TIMS and SIMS calibration errors (or bias) are now present in the Monte Carlo program.
- Measurement error parameters used to describe TIMS and SIMS have been updated with current best estimates.
- The U236/U235 ratio has been eliminated from TIMS measurement suite as redundant.
- Modeling of the P-search analysis steps in the SIMS particle measurement procedure have been eliminated; The error analysis now assumes that SIMS produces a single ratio at a sampling location with a known uncertainty.

- Data describing the generic reactor used in this study have been changed.
- The estimation procedure has been simplified; Lithium ratios have been removed from the measurements and all robust statistical steps have been eliminated from the estimation procedures.

The benefit of the robust steps depends upon the severity of sample contamination. Because of all the work being expended on quality control and sample handling, contamination is expected to be relatively modest, and the biases/errors originating from contamination are not large relative to other errors, even for "non-robust" estimation procedures.

- The global fluence model, which is used to produce an estimate of the reactor's 3D fluence field, has been improved.

We include an analysis of various reactor sample plans (i.e. prospective locations for sampling). The objective is to find small sets of locations that will produce total Pu estimates with the smallest standard error possible.

Scenarios have been added that quantify the effect of calibration error and uncertainty in the global fluence field model.

#### **References**

Talbert RJ, Reid BD, Heasler PG, Gerlach DC, Heeb CM, Pauley KA, "Accuracy of Plutonium Production Estimates from Isotopic Ratios in Graphite Reactors," PNL-RTC-0693-REV1-DEL, Feb. 1995.

Reid BD, Gerlach DC, Heasler PG, Livingston JV, "Trawsfynydd Plutonium Estimate," PNNL, Sept. 1997.