Isotope Ratio Method Analysis of the Ford Nuclear Reactor

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Abstract

The Isotope Ratio Method (IRM) is a technique for estimating the energy production in a fission reactor by measuring isotope ratios in non-fuel reactor components. The isotope ratios in these components can then be directly related to the cumulative energy production with standard reactor modeling methods.

All reactor materials contain trace elemental impurities at parts per million levels, and the isotopes of these elements are transmuted by neutron irradiation in a predictable manner. While measuring the change in a particular isotope's concentration is possible, it is difficult to correlate to energy production because the initial concentration of that element may not be known. However, if the ratio of two isotopes of the same element can be measured, the energy production can then be determined without knowing the absolute concentration of that impurity since the initial natural ratio is known. This is the fundamental principle underlying the IRM. Extremely sensitive mass-spectrometric methods are currently available that allow accurate measurements of the impurity isotope ratios in a sample. Additionally, indicator elements with stable activation products have been identified so that their post-irradiation isotope ratios remain constant.

This method has been successfully demonstrated on graphite-moderated reactors. Graphite reactors are particularly well-suited to such analyses since the graphite moderator is resident in the active core for the entire period of operation. Applying this method to other reactor types is more difficult since the resident portions of the reactor structure available for sampling are typically outside the active core. The goal of this research is to evaluate whether the IRM can produce meaningful results for water-moderated research reactors.

In this paper, we use the IRM to estimate the energy production of the University of Michigan's Ford Nuclear Reactor (FNR) from 1992 until its shutdown in 2003. The FNR is a 2MW, swimming-pool reactor that is currently being decommissioned. In 1992, several ex-core structural components were added to support a large irradiation program. We have taken samples from some of these components, analyzed them and produced an estimate of the total energy production. Our preliminary results are in good agreement with the actual operating history of the reactor during that time period.

Introduction

The measurement of characteristic radiation from fission products or activation products has long been used to infer neutron fluence for a wide variety of applications. Only recently, however, have mass spectrometric methods capable of measuring isotope ratios of extremely low concentration impurity elements become available. In the mid-1990s, an effort was undertaken at the Pacific Northwest National Laboratory to develop a technology for verifying plutonium production in graphite-moderated reactors [, , , and]. The technique developed, known as the Graphite Isotope Ratio Method (GIRM), was based on the fact that neutrons produced from fission reactions are also parasitically absorbed in non-fuel components such as reactor structural material, control rods, coolant, and the graphite moderator. The fluence in these non-fuel components is proportional to the total number of fissions that have occurred in the fuel. This, in turn, can be directly related to the total plutonium production in the reactor. Therefore, if the fluence in the graphite can be determined, the cumulative plutonium production can be inferred.

Even the highest purity, reactor-grade graphite contains elemental impurities at parts per million levels and the isotopes of these elements are transmuted by neutron irradiation in a predictable manner. While measuring the change in a particular isotope's concentration is possible, it is difficult to correlate to energy or plutonium production because the initial concentration of that element may not be known. However, if the ratio of two isotopes of the same element can be measured, the fluence can then be determined without knowing the absolute concentration of that element, since the initial ratio is simply the natural ratio. This is the fundamental principle underlying the Graphite Isotope Ratio Method. A key advantage of this method compared to more traditional activation analysis is that numerous indicator elements with stable activation products have been identified so that their isotope ratios remain constant after irradiation.

Graphite reactors possess several characteristics that make this method particularly appealing. The concentrations of key impurity elements are so low that they do not perturb the spatial or energy flux shape in the graphite – that is, they are infinitely dilute. The underlying physics of graphite reactors results in flux profiles that are relatively smooth. Most importantly, the graphite moderator is resident in the core for the entire lifetime of the reactor – it provides a permanent record of reactor operation. This method has been successfully demonstrated [] and can produce plutonium production estimates with a root mean standard error of approximately two percent.

The goal of our current work is to evaluate the effectiveness of this technique for water moderated research reactors. Since the structural components of such reactors available for sampling are mainly outside of the active fuel, the analysis is more complicated. We hope, however, to show that meaningful results can be obtained.

The primary application for the earlier work has been non-proliferation treaty verification. As such, "meaningful results" can mean a variety of things. In the case of graphite production reactors, the goal has been to estimate the cumulative plutonium produced in a reactor to within a few percent. In the case of research reactors, simply confirming declared operations in a broad sense and detecting relatively major deviations, such as the large scale replacement of reflector material with plutonium or tritium producing targets.

There are other possible applications for this technology. One of the most appealing is confirming the burnup of spent light water reactor fuel. By analyzing very small samples from each grid strap location, the axial burnup profile of a fuel assembly can be accurately determined. Thus far, we have verified that suitable indicator elements are present in measurable quantities in the zircaloy samples we have studied.

In the remainder of this paper, we will describe the Ford Nuclear Reactor, discuss our sampling and measurement campaign, review the preliminary IRM modeling effort, present preliminary results and provide some conclusions and our plans for future work.

The Ford Nuclear Reactor

The Ford Nuclear Reactor operated on the campus of the University of Michigan from 1957 until 2003. It is a 2MW thermal, pool type reactor that utilized MTR fuel. The reactor was heavily used for training and research until its shutdown in 2003. In 1992, several modifications were made in the reactor pool including the installation of two aluminum guard plates that protected and isolated the core from the experiments in the south and east experimental grids. The south and east plates are Aluminum 6061-T6 and were resident in the core for approximately 4800 MWd. Figure 1 shows a top view of the FNR core and the south and east experimental grids. 21 samples were taken from the south guard plate and 17 from the east guard plate (though, to date, only 5 samples from the south guard plate have been analyzed).

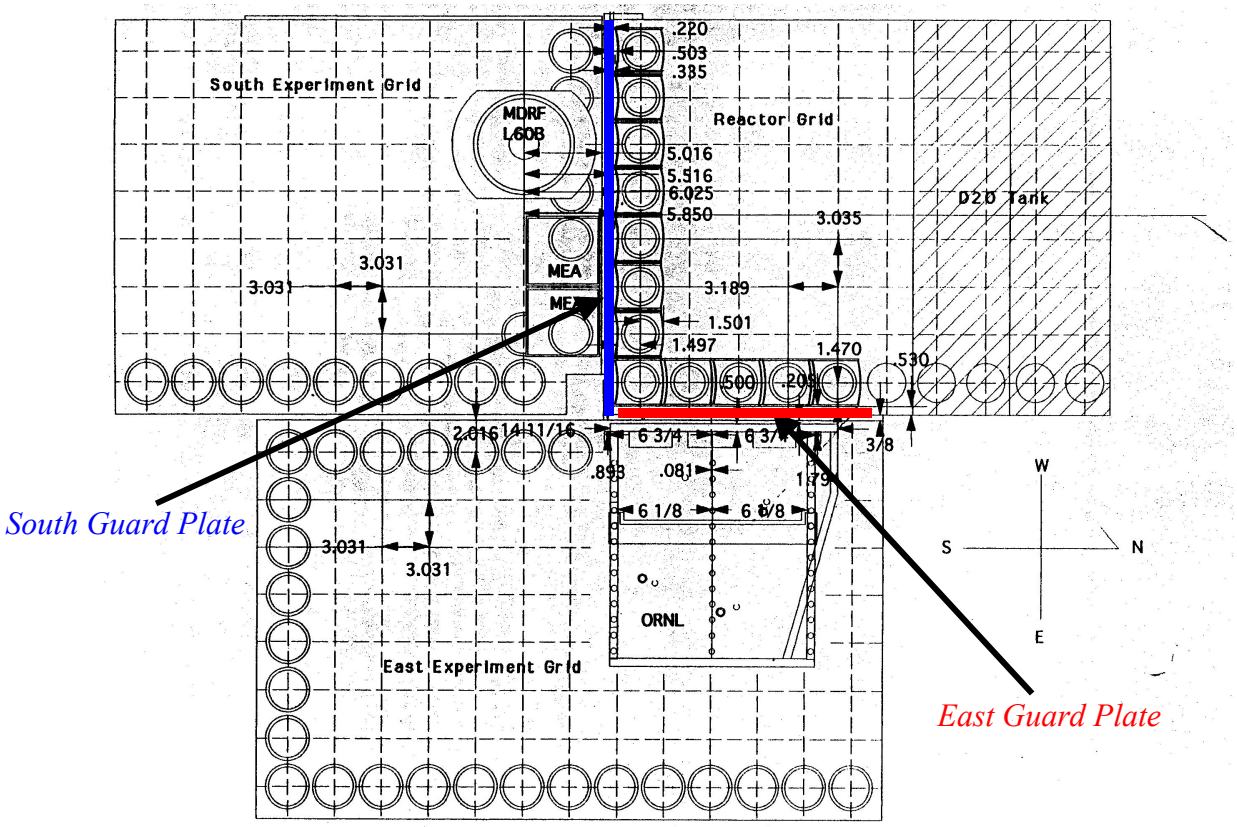

Figure 1 – FNR core/experimental grid layout (top view)

Indicator Elements

The first step in an IRM analysis is to identify suitable indicator elements. Indicator elements should exist in sufficient quantities to be accurately measured, have stable activation products, and have cross-sections of suitable magnitude to result in meaningful isotope ratio changes for the fluence range in question. For high-fluence reactors, the Ti^{48}/Ti^{49} ratio has been used successfully. For low-fluence measurements, the B^{10}/B^{11} ratio is an excellent indicator element. The Cl^{36}/Cl^{35} ratio has been effective for intermediate fluence measurements. A wide variety of uranium and plutonium ratios are appropriate for both low- and high-fluence applications. The use of multiple indicator elements, such as boron and chlorine, tends to reduce the overall uncertainty associated with an IRM analysis. Table 1 summarizes the properties of several useful

indicator elements. The applicable 2200m/s cross sections [] are shown in parenthesis for all but the U/Pu isotopes, since their ratio changes cannot be characterized by a single cross section.

Table 1 – IRM Indicator Elements

Element	Key Isotope Ratios	Fluence Range
Boron	B^{10}/B^{11}	Low (3838b)
Lithium	Li^{6}/Li^{7}	Low-Intermediate (941b)
Chlorine	Cl^{36}/Cl^{35}	Intermediate (43.6b)
Titanium	Ti^{48}/Ti^{49}	Intermediate-High (7.9b)
Uranium	U^{235}/U^{238}, U^{236}/U^{238}	Low-High
Plutonium	Pu^{240}/Pu^{239}, Pu^{241}/Pu^{239}, Pu^{242}/Pu^{239}	Low-High

Sampling, Preparation and SIMS Measurement

Recently, FNR staff moved the south and east guard plates from the reactor pool to their hot cell facility where a series of samples, approximately 3/8 inch in diameter, were cut with plug cutting tool. 21 samples were taken from the south plate and 17 from the east plate. Turnings were also collected from several of the samples. The contact dose rate of the samples was approximately 20-60 mR/hr. All the samples have been received by PNNL for Secondary Mass Spectrometry (SIMS) isotope ratio analysis.

The samples were prepared for analysis in a glovebox where slices were cut from the cylindrical slugs and a variety of cleaning methods applied. Previous experience with graphite samples has indicated that surface contamination can be a serious issue, particularly with natural boron. This is largely because of the high porosity of graphite. The experience so far with aluminum is that surface contamination is not a serious issue.

Both the boron and chlorine appear to occur in inclusions within the aluminum matrix. Since the observed concentrations of boron and chlorine are so low, approximately one day of analysis has been required to obtain a single isotope ratio measurement with satisfactory statistics. The actual bulk concentration of these impurities is difficult to quantify since they occur in inclusions. However, both are probably present at sub-ppm levels.

Reactor Modeling and Energy Production Estimates

In order to estimate the energy production of the reactor, the indicator element reaction rates must be calculated in each sample location. The actual operation of the FNR was characterized by frequent refueling and fuel shuffling, and a wide variety of both in-core and ex-core experiments. Because of this complexity, our preliminary efforts have simply used a characteristic fuel loading scheme with fresh fuel so that the flux in the aluminum guard plates is constant. The three-dimensional diffusion code 3DB [] was used along with 89-group NJOY [] generated cross sections to calculate the boron and chlorine reaction rates for this core configuration. The measured isotope ratios at each sample location can then be related to the energy production.

Clearly, the uncertainty associated with the energy production estimate is strongly dependent on having suitable calculational tools and detailed reactor design/operational data so that accurate reaction rates can be calculated at each sample location. Ideally, the isotope ratios at each location will correspond to the same

total energy production. The calculated relative flux profile in the FNR core is shown in Figures 2a and 2b. Note that the flux is roughly symmetric across the south guard plate.

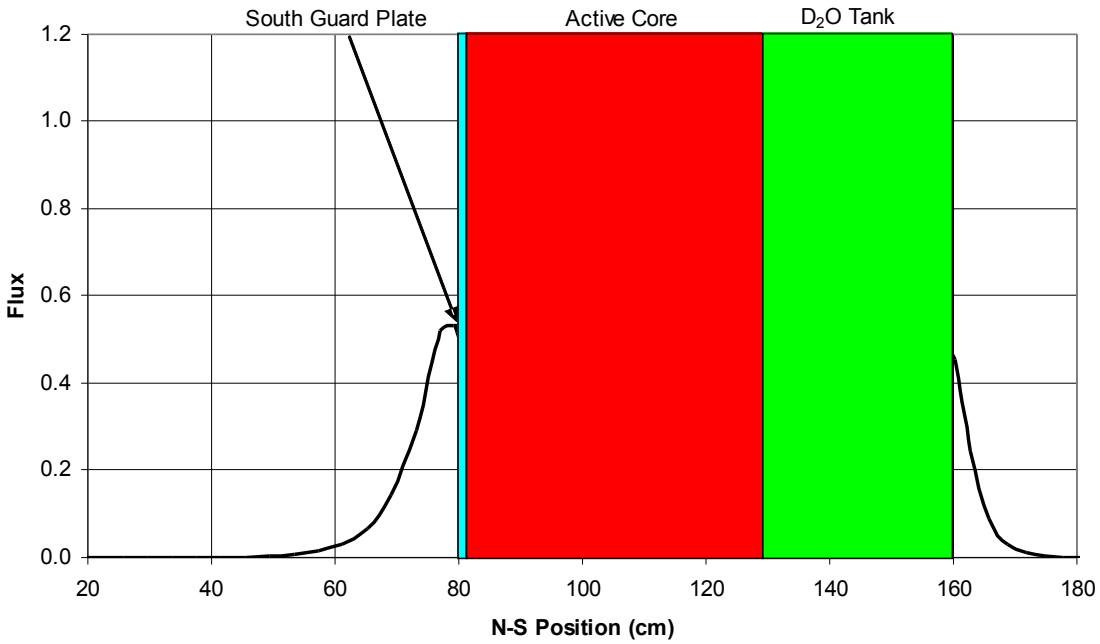

Figure 2a – FNR relative flux profile (N-S)

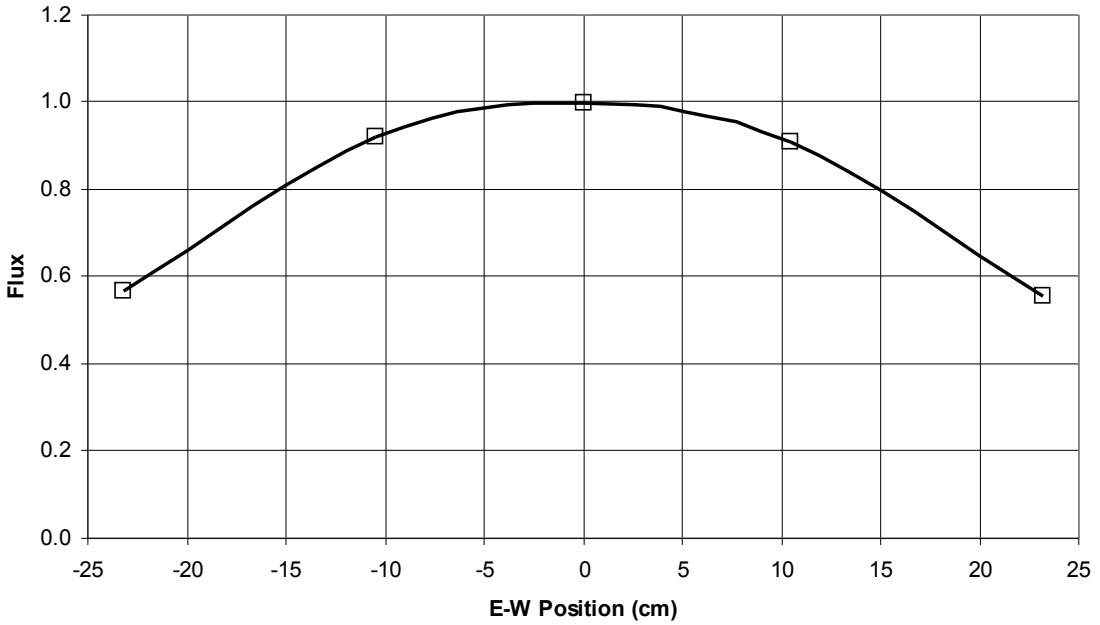

Figure 2b – FNR relative flux profile (E-W along the south guard plate)

The necessity of fairly detailed 3D calculations for the FNR is in sharp contrast to a graphite-moderated reactor. Since samples can be taken directly from the active core region, local plutonium production estimates can be made with simple pin-cell calculations. These local estimates can then be combined into a global production estimate using regression. We have successfully used functions from the eigenfunction solution to the diffusion equation as the basis of the regression. However, our experience indicates that

using some calculated 3D flux shapes in regression basis function set can reduce the overall uncertainty. Additionally, the flux shape in a graphite reactor tends to be relatively smooth and the operational parameters are much more restrictive so that even without detailed design and operational data, accurate estimates can be made.

While several studies have evaluated the uncertainty in GIRM calculations [,], similar work has not yet been completed for this analysis. For graphite reactors, approximately 100 carefully selected samples can produce global plutonium production estimates with root mean standard errors of less than 2%. The major sources of error include sample position/orientation uncertainties, mass spectrometry measurement uncertainties, cross-section and calculational uncertainties, and uncertainties associated with the reactor design and operational parameters. Our expectation is that while the uncertainty in the total energy production estimate for water moderated research reactors will be significantly larger than is possible in graphite reactors, it will still be relatively low – perhaps in the range of 10-20%.

Preliminary Results and Discussion

Five samples from the south guard plate were been analyzed for boron and chlorine ratios. B^{10} is consumed mainly through its (n,α) reaction while B^{11} remains essentially constant. As fluence increases, the B^{10}/B^{11} ratio falls from its natural value of 0.248. Cl^{36} is produced by an (n,γ) reaction from Cl^{35}, so the ratio begins at zero and rises with fluence. This behavior is shown in Figure 3. In these samples, the B^{10}/B^{11} ratio has fallen two orders of magnitude and is nearing the end of its useful range as an indicator element. Not only does the B^{10} signal become lower during the SIMS measurement, the effect of any natural boron contamination is amplified since most of the B^{10} originally in the aluminum has been consumed.

Figure 3 shows the measured isotope ratios for one of the samples. The left vertical axis indicates B^{10}/B^{11} ratio and the right vertical axis indicates Cl^{36}/Cl^{35} ratio as a function of MWd. Both the boron and chlorine isotope ratios indicate a total energy production of just under 4800 MWd.

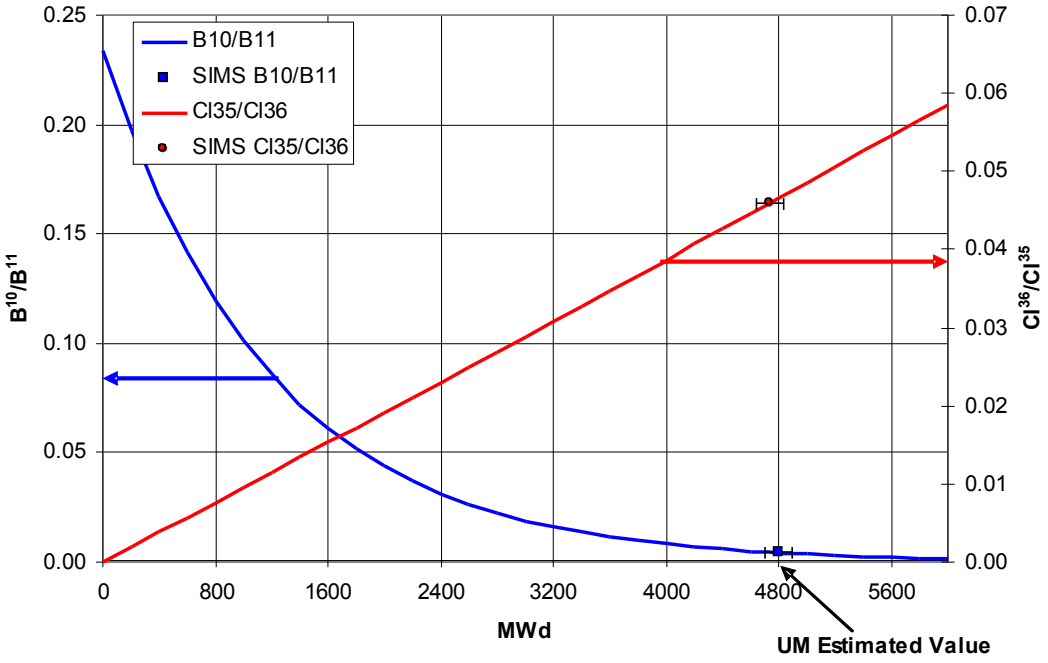

Figure 3 – B^{10}/B^{11} and Cl^{36}/Cl^{35} as a function of MWd at -23.2cm

The five samples that have been analyzed were taken across the south guard plate, approximately 6 inches below the top of the fuel. The sample position relative to the E-W center of the south guard plate (see Figure 1) and the measured isotope ratios and their standard deviations are shown in Table 2. Additionally, the 3DB results for the corresponding energy (MWd) required to achieve that ratio at each position is shown.

Table 2 – Measured Isotope Ratios and Calculated Energy Production

Position (cm)	B^{10}/B^{11} (σ)	B^{10}/B^{11} MWd	Cl^{36}/Cl^{35} (σ)	Cl^{36}/Cl^{35} MWd
-23.2	4.8E-03 (8.5E-05)	4794	4.6E-02 (7.9E-04)	4741
-10.5	5.2E-04 (1.4E-05)	5231	7.3E-02 (2.2E-03)	5223
0.0	1.6E-04 (4.8E-06)	5762	9.0E-02(9.7E-04)	5915
10.5	2.2E-04 (5.5E-06)	6014	8.4E-02 (1.9E-03)	6044
23.2	2.2E-03 (5.2E-05)	5881	5.8E-02 (1.0E-03)	6060

The values agree reasonably well with the actual estimated energy production of 4800 MWd. In particular, note that the relative boron-chlorine energy estimates are consistent. This is compelling evidence that the isotopic ratios are measured accurately and that no significant contamination is present. These values are shown graphically in Figure 4.

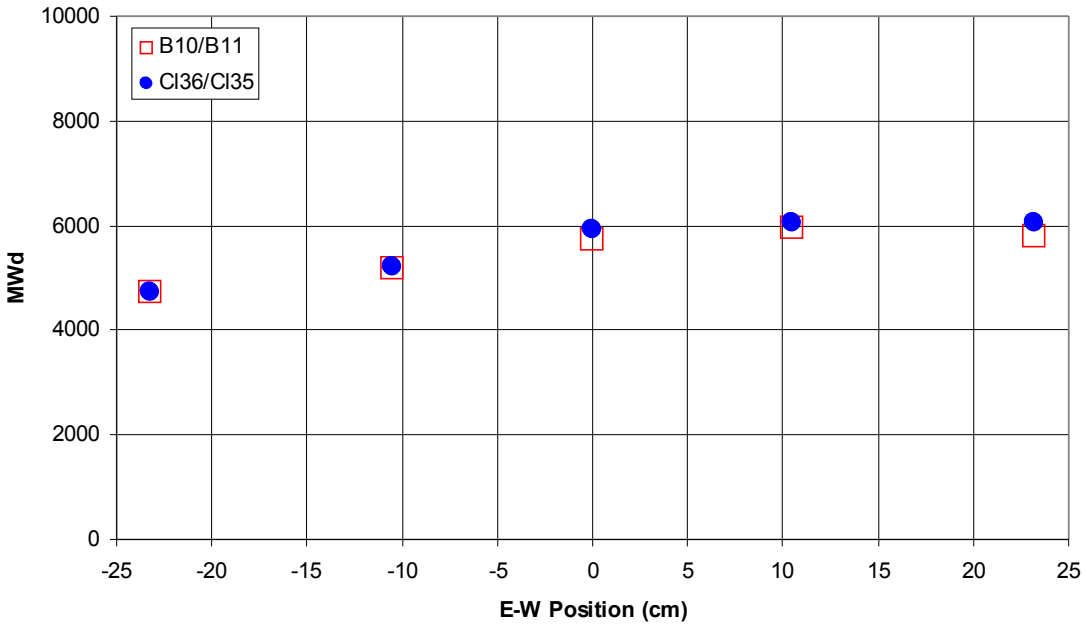

Figure 4 – Estimated Energy Production (E-W along the south guard plate)

Unlike our simple reactor physics model, the actual energy production is not symmetric about the middle of the south guard plate. Our current model does not presently account for a number of operational and experimental design features known to have existed. In this case, the most important feature is that the core was loaded in such a way that the power tended to peak towards the east side of the core. In all likelihood, this accounts for the observed tilt in energy production toward the east.

Conclusions and Future Work

While our preliminary results are satisfactory, we hope to refine our reactor models and analyze additional samples to produce a more accurate estimate. Though the boron and chlorine ratios on the east side of the plate yield energy production estimates about 25% higher than the known value, correcting the physics models should reduce those errors significantly. Additionally, as more samples are analyzed we anticipate further improvements. Of course, the possibility of a systematic bias associated with the SIMS measurements or the calculations cannot be ruled out. Regardless, these first results are quite encouraging and the flux tilt toward the east side of the core was easily detected. We do intend to perform a detailed uncertainty analysis so that the actual standard error of the estimate can be bounded.

Recall that our reactor calculations have been performed with a diffusion theory code. While this is more than adequate for large, graphite-moderated reactors, in this case we have a small reactor with steep flux gradients and are primarily interested in reaction rates at the core-reflector interface. Diffusion theory is known to have inaccuracies for such problems. Therefore, we plan to perform some additional core analysis with the 3D discrete ordinates code Attila []. Additionally, we hope to identify a suite of indicator elements that could provide spectral information.

Finally, we hope to test this method on operating research reactors. This, of course, would require a non-destructive sampling tool that could be remotely operated in a pool. We hope to test several sampling tool concepts on the FNR reactor grid and, if possible, on other reactor types.

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