Isotope Ratio Method Analysis of the Ford Nuclear Reactor

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Isotope Ratio Method Basics

- A very simple, mass spectrometry based technique for determining fluence in an irradiated sample. Fluence, in turn, can be related to the total energy production in a reactor.
- Originally developed for non-proliferation applications (estimation of plutonium production in graphite-moderated reactors).

Isotope Ratio Method Basics

- Basis – The isotopes of trace elements found in reactor materials are transmuted during neutron irradiation. If the ratio of isotopes from a trace element can be measured, the fluence can be inferred even if the absolute concentration is unknown.

- Goal – Determine whether this method can produce meaningful results for water moderated research reactors.

Applications

- **Non-proliferation** – *verification* of declared operations and/or detection of significant design/operational deviations (such as the replacement of reflector materials with plutonium or tritium producing targets or the use of enriched fuel)
- **Burnup credit** – confirmation of axial exposure in LWR fuel assemblies
- **Neutron fluence measurement** in reactor materials
- **Code validation**

Pros and Cons

- Pros
  - Stable isotope ratios can be measured any time after irradiation
  - Accurate and tamper resistant
  - Our primary mass spectrometry method is basically non-destructive
- Cons
  - In-situ measurements difficult (if not impossible)
  - Contamination (boron in particular) must be addressed
  - Future operability of reactor constrains sampling method and locations

Methodology

- Identify indicator elements based on expected fluence
- Take samples
- Measure key isotope ratios
  - SIMS for low Z materials
  - TIMS for high Z materials
- Produce estimate
  - Reactor calculations to relate key parameter (energy or plutonium production) to isotope ratios in sample locations
    - Relatively simple for graphite reactors (Trawsfynydd example)
    - More difficult for research reactors – more design/operational information required to achieve comparable accuracy
  - Uncertainty/error analysis

Indicator Elements

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

IRM

- For example:

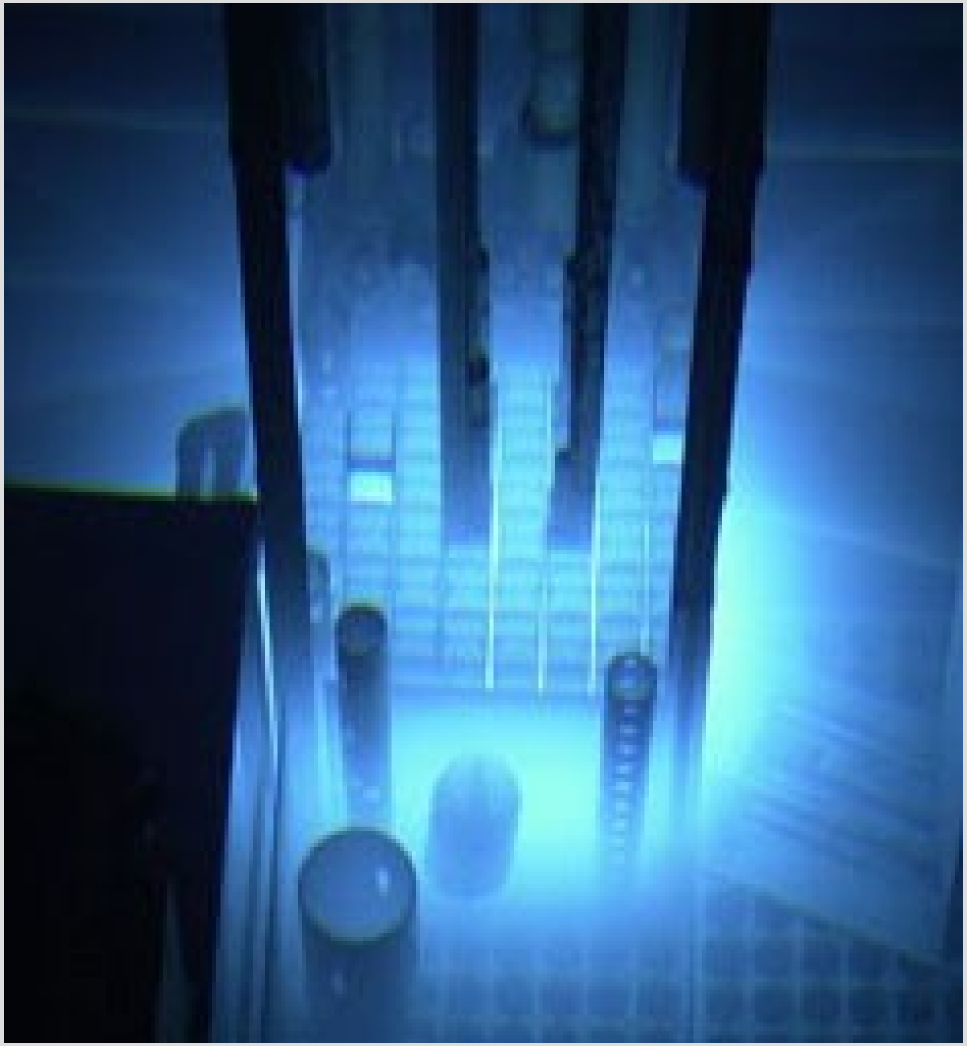
$$\frac{^{10}B}{^{11}B} = 0.248e^{-\sigma_{10}\phi t}$$

$$\frac{^{36}Cl}{^{35}Cl} = e^{-\sigma_{35}\phi t} - 1$$

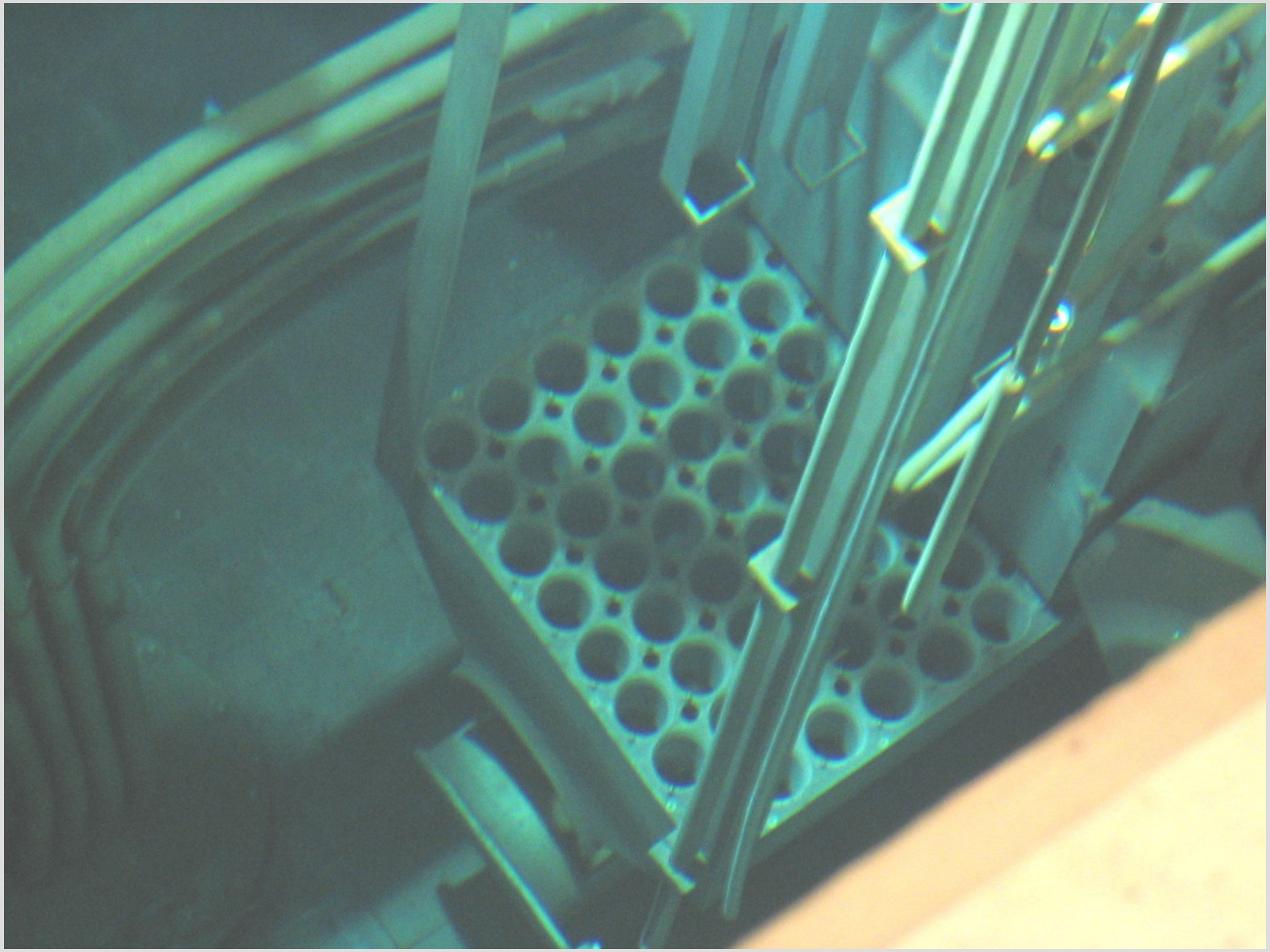
Ford Nuclear Reactor (FNR)

- Located on the campus of the University of Michigan
- 2MW, MTR-fuel, numerous beam ports and in/ex-core experimental locations.
- Timeline:
  - Initial criticality – 1957
  - Shutdown – 2003
  - Currently being decommissioned
  - South and East Guard Plates added during the 1991-92 operating year (approximately 4800 MWd of operation since then)

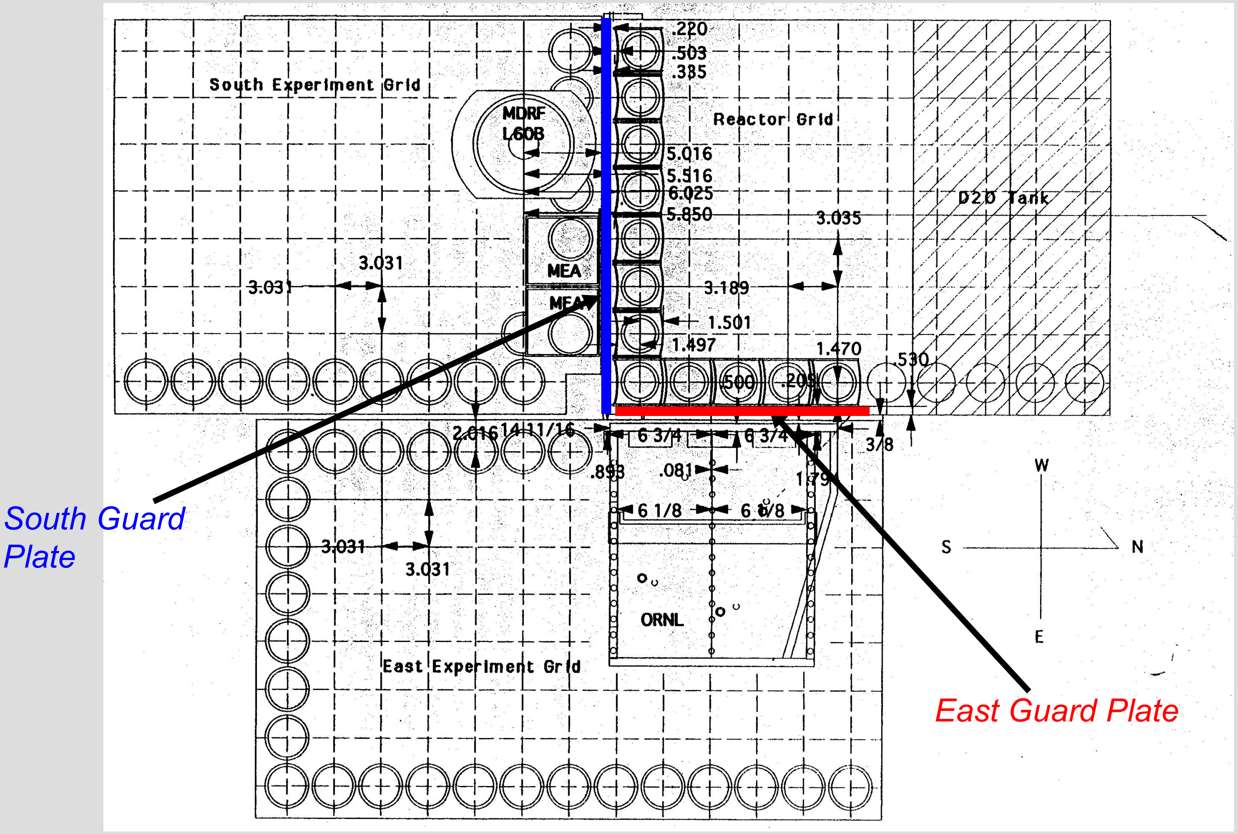
FNR



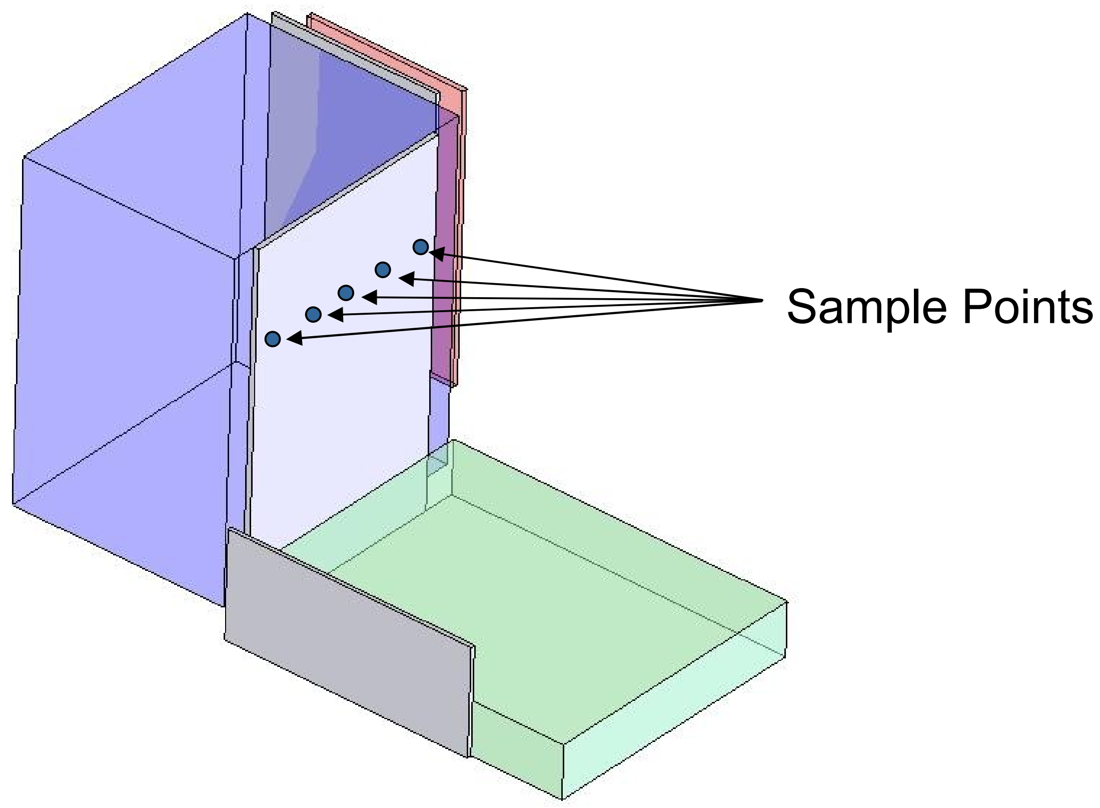
Reactor Grid



FNR Experimental Grids

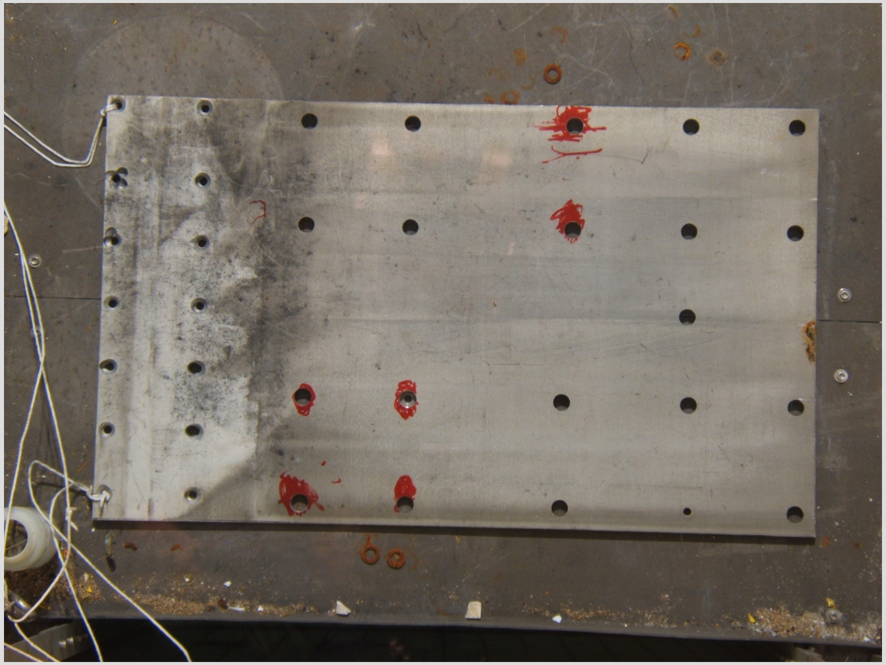


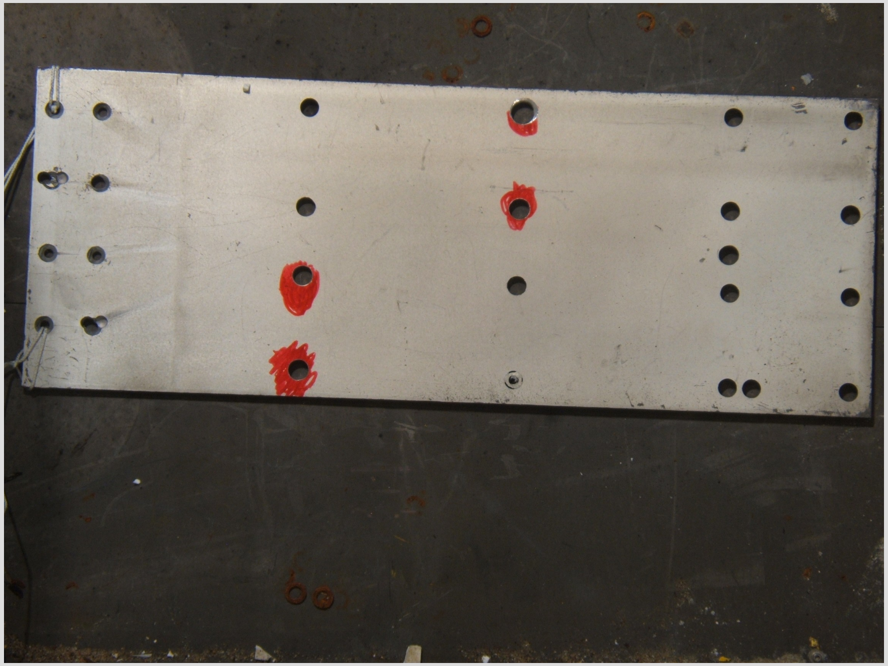
FNR SW View



Sampling in the Phoenix Lab Hot Cell

South and East Guard Plate Sample Locations

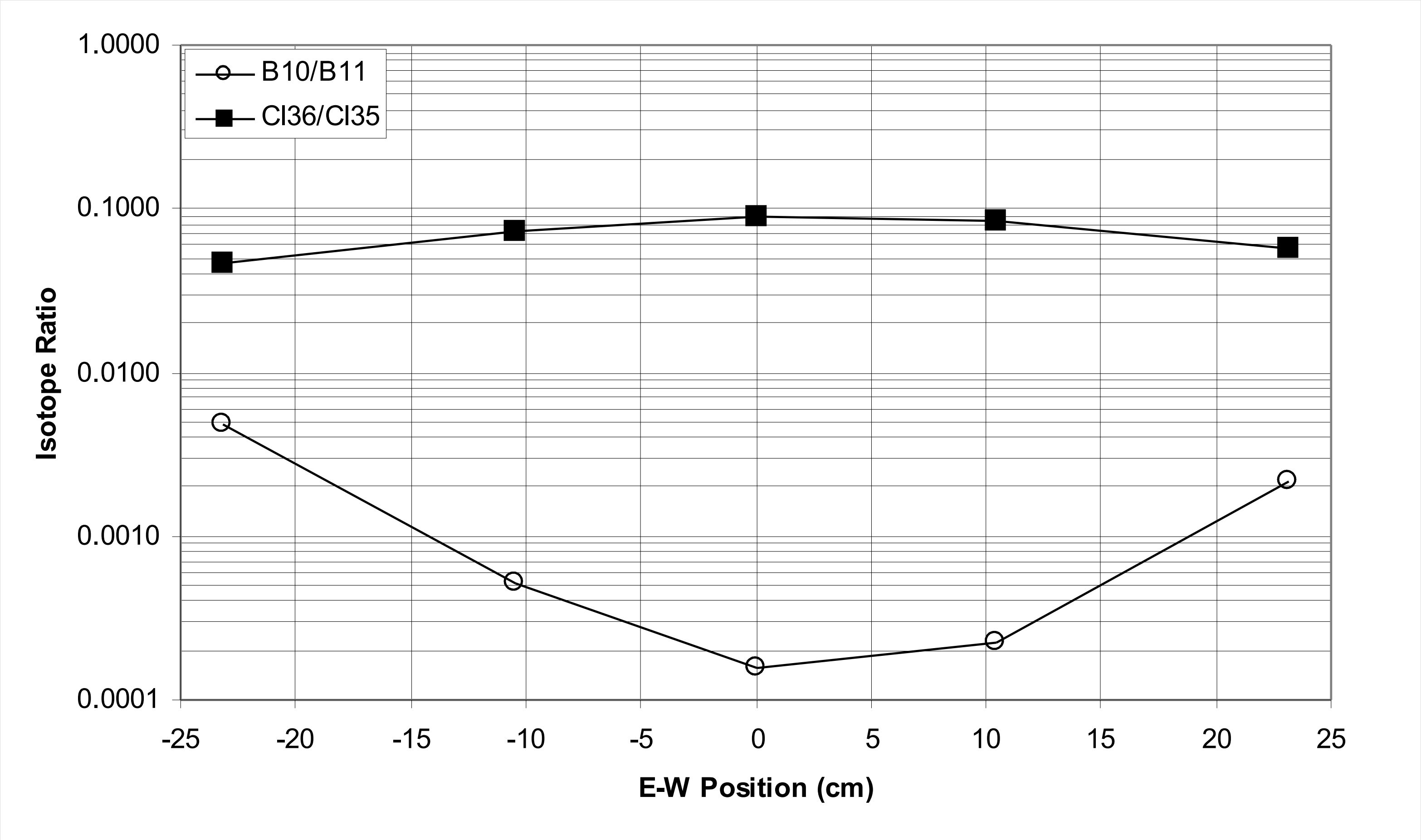




Preliminary SIMS Results

Position (cm)	$^{10}B/^{11}B$	σ	$^{36}Cl/^{35}Cl$	σ
-23.2	4.8E-03	8.5E-05	4.6E-02	7.9E-04
-10.5	5.2E-04	1.4E-05	7.3E-02	2.2E-03
0.0	1.6E-04	4.8E-06	9.0E-02	9.7E-04
10.5	2.2E-04	5.5E-06	8.4E-02	1.9E-03
23.2	2.2E-03	5.2E-05	5.8E-02	1.0E-03

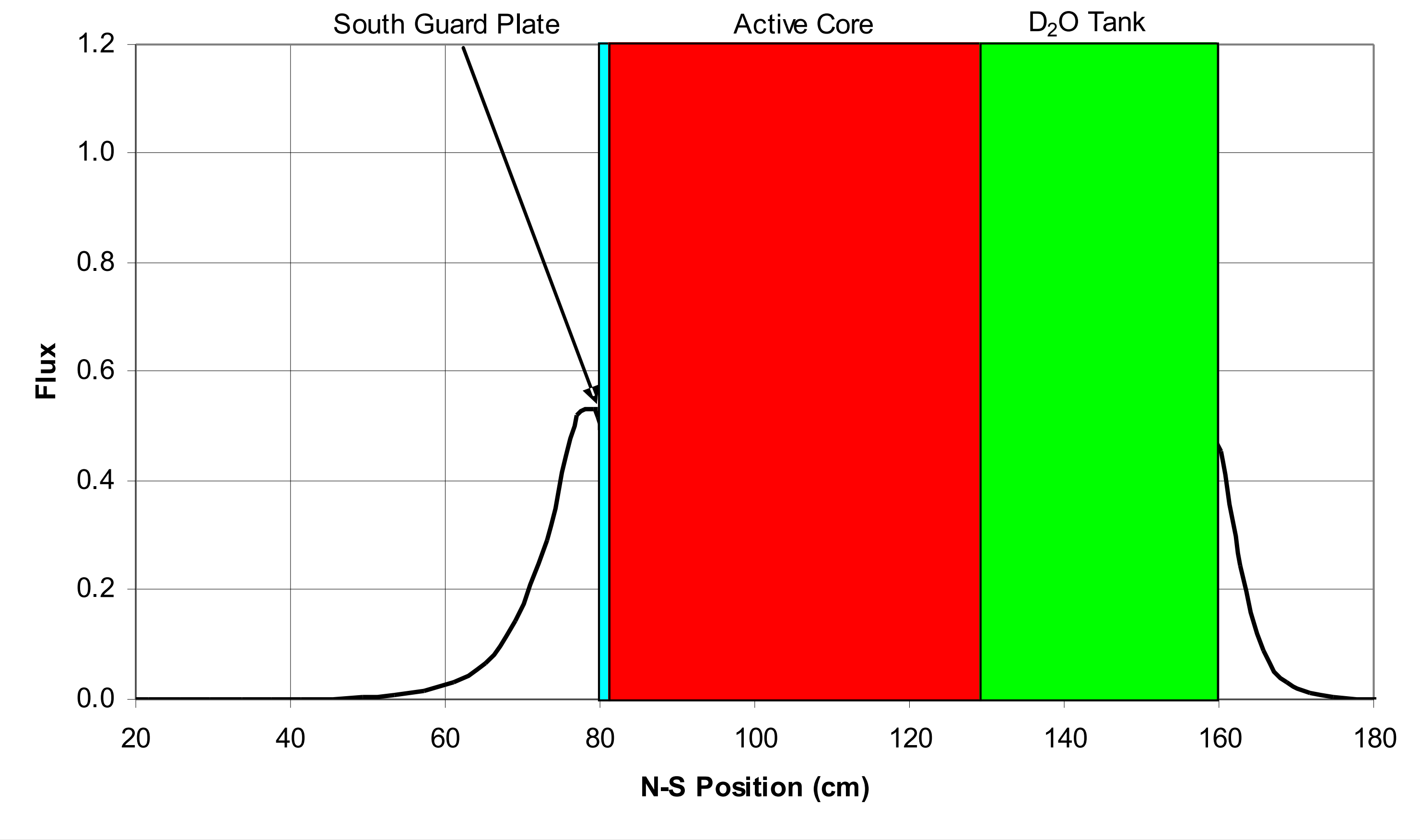
Measured Isotope Ratios



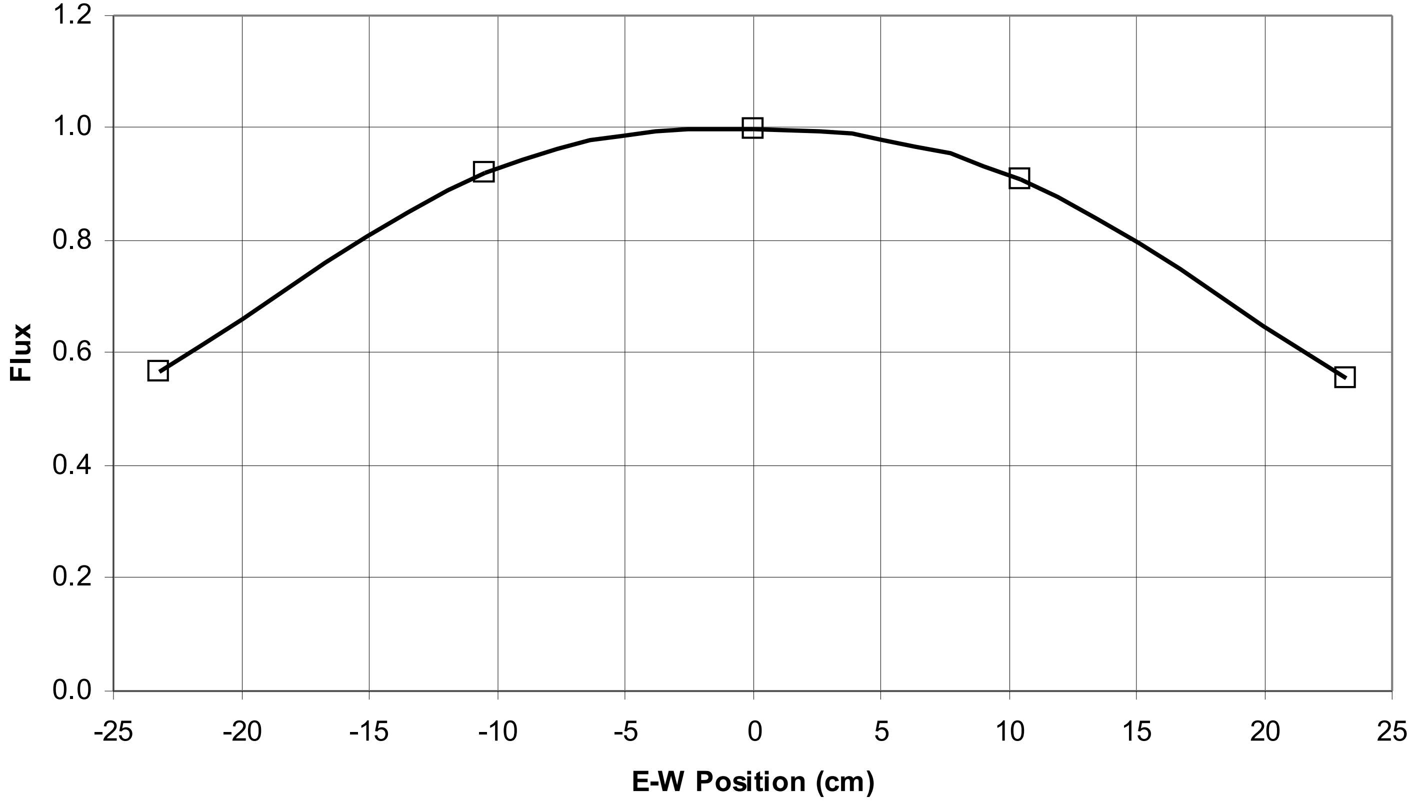
3DB Model

- Reactor modeling
  - 3DB (finite difference diffusion)
  - NJOY (89 group cross sections)
- The current model does not explicitly account for the known fuel loading scheme (fuel loaded to keep the flux as constant as possible along the East Experiment Grid)
- Diffusion theory struggles at material interfaces and for small, high flux gradient systems.

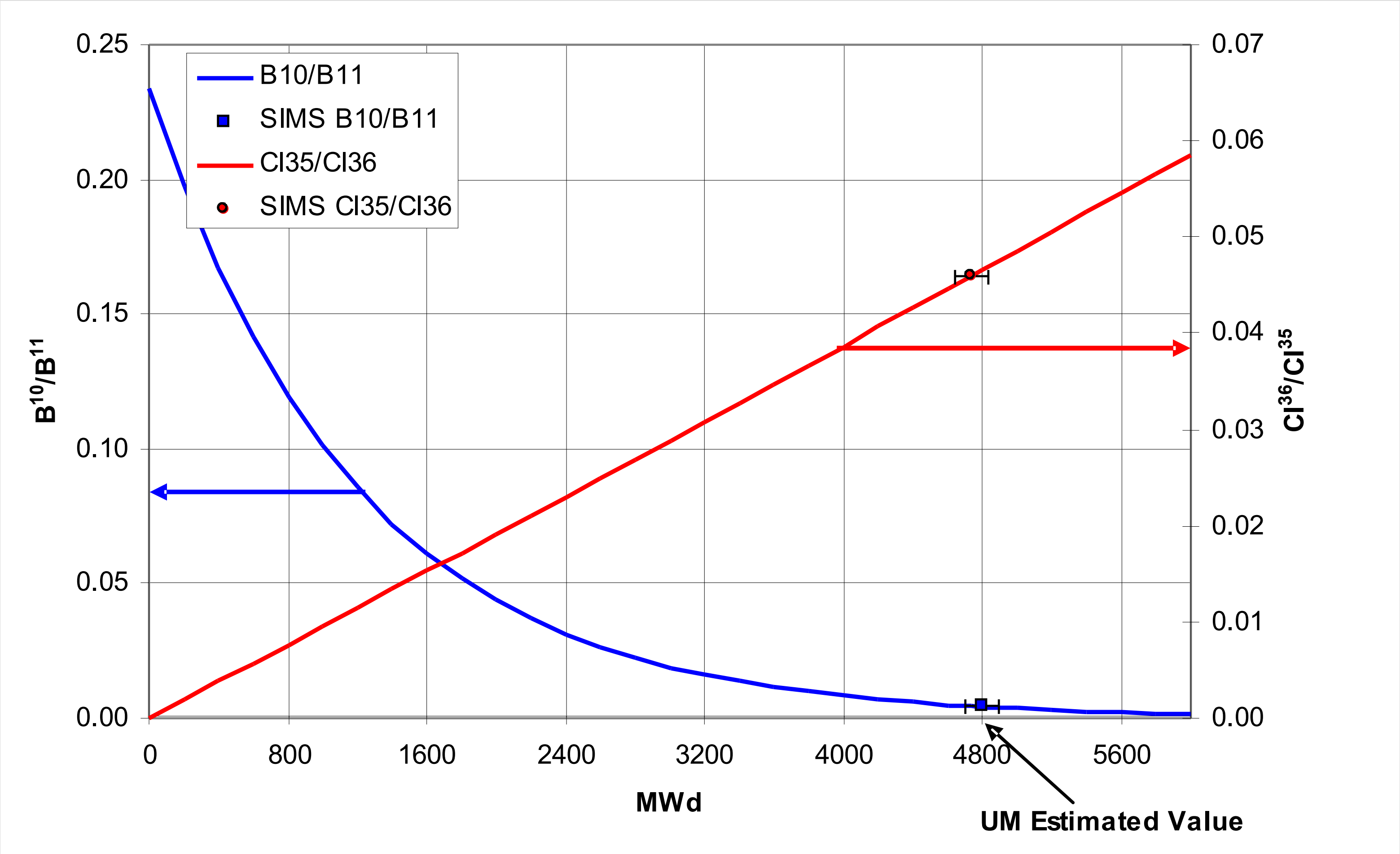
FNR 3DB Calculated Flux Profile



FNR 3DB Calculated Flux Profile



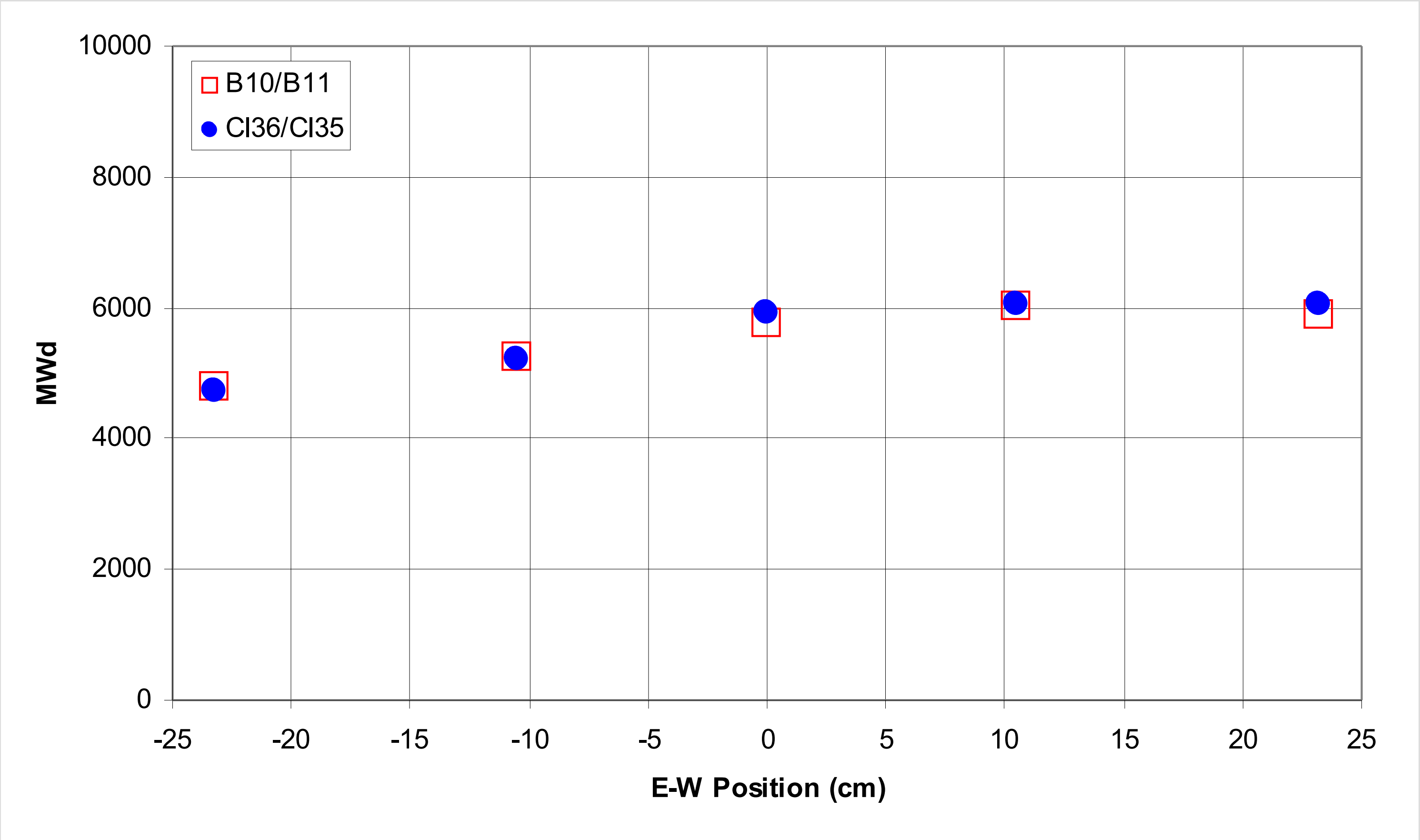
Preliminary Results (E-W=-23.2cm)



Energy Estimates at Sample Locations

Position (cm)	$^{10}B/^{11}B$ MWd	$^{36}Cl/^{35}Cl$ MWd
-23.2	4794	4741
-10.5	5231	5223
0.0	5762	5915
10.5	6014	6044
23.2	5881	6060

Preliminary Results



Preliminary Results

- These results clearly show that more operational detail is needed in our reactor model.
- The data can be used to help refine the reactor model or reveal unknown design/operational features
- Ultimately, self-consistent results should be obtained.

Preliminary Results

- Detailed uncertainty and error analysis not yet complete.
  - For graphite reactors, RMSEs of less than 2% are expected and have been observed in actual tests.
  - In this case, because the complex nature of FNR's operation, we anticipate RMSEs of 10-20%
- The average of the estimated energy from the five samples is 5570 MWd – about 16% high.
- By improving the model and analyzing more samples, we expect to reduce the RMSE.

Future Work

- Improve physics models (use of more accurate operational data and analysis with Attila/WIMS)
- Develop a remote sampling tool to test on FNRs reactor grid
- Test on other research reactor types (TRIGA, IRT, etc.)