Integral Testing of the ^{239}Np Capture Cross Section using Reactor Experiments

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Validation Committee
Brookhaven National Laboratory

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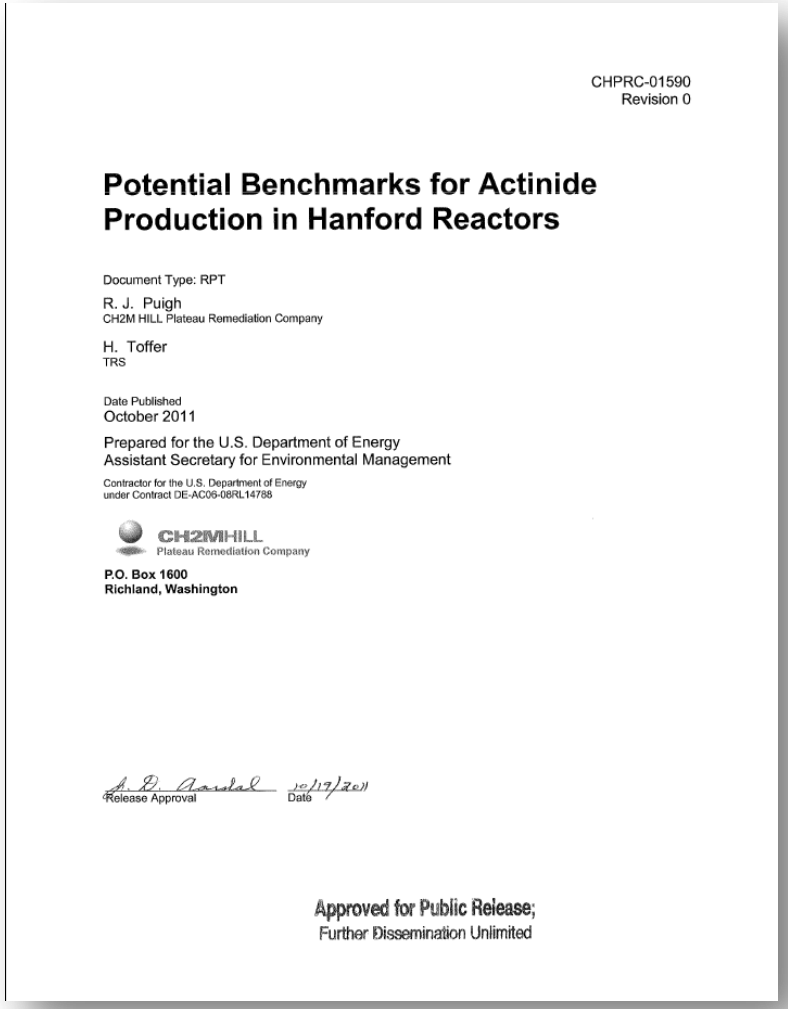
Introduction

- Reactor benchmarks involving isotopic measurements of irradiated fuel have provided important integral data validation (Cs, Sm, Eu)
- In mid-to-late 1990s, many of the primary reports from Hanford reactor experiments conducted in 1960s were declassified
- Experimental data from radiochemistry on discharged fuel elements was measured as the basis for predictive capability at the time of early production reactors, given limited nuclear data and simulation methods available
- In 2010, the DOE Nuclear Criticality Safety Program (NCSP) initiated a multi-laboratory effort to investigate potential use of the historical measurement data from Hanford and Savannah River reactors
- NCSP objective is to compile and review data for potential use as benchmarks for improved integral actinide cross section data for the criticality safety program

Hanford Reactor Benchmarks



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Potential Benchmarks for Actinide Production in Hanford Reactors

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- Hanford experiments evaluated by DOE-EM for criticality safety program (B, N, and K reactors)
- Nine experiments identified as having sufficient documentation to be used as potential benchmarks
- Experiments performed in 1967-1970
- Declassified in 1995 for public release
- Destructive analysis data from high precision mass spectrometry
- Very low exposures
  - Commercial LWR: ~15,000 – 70,000 MWd/t
  - CANDU Nat U: ~7,500 MWd/t
  - Hanford: 285 – 1,720 MWd/t

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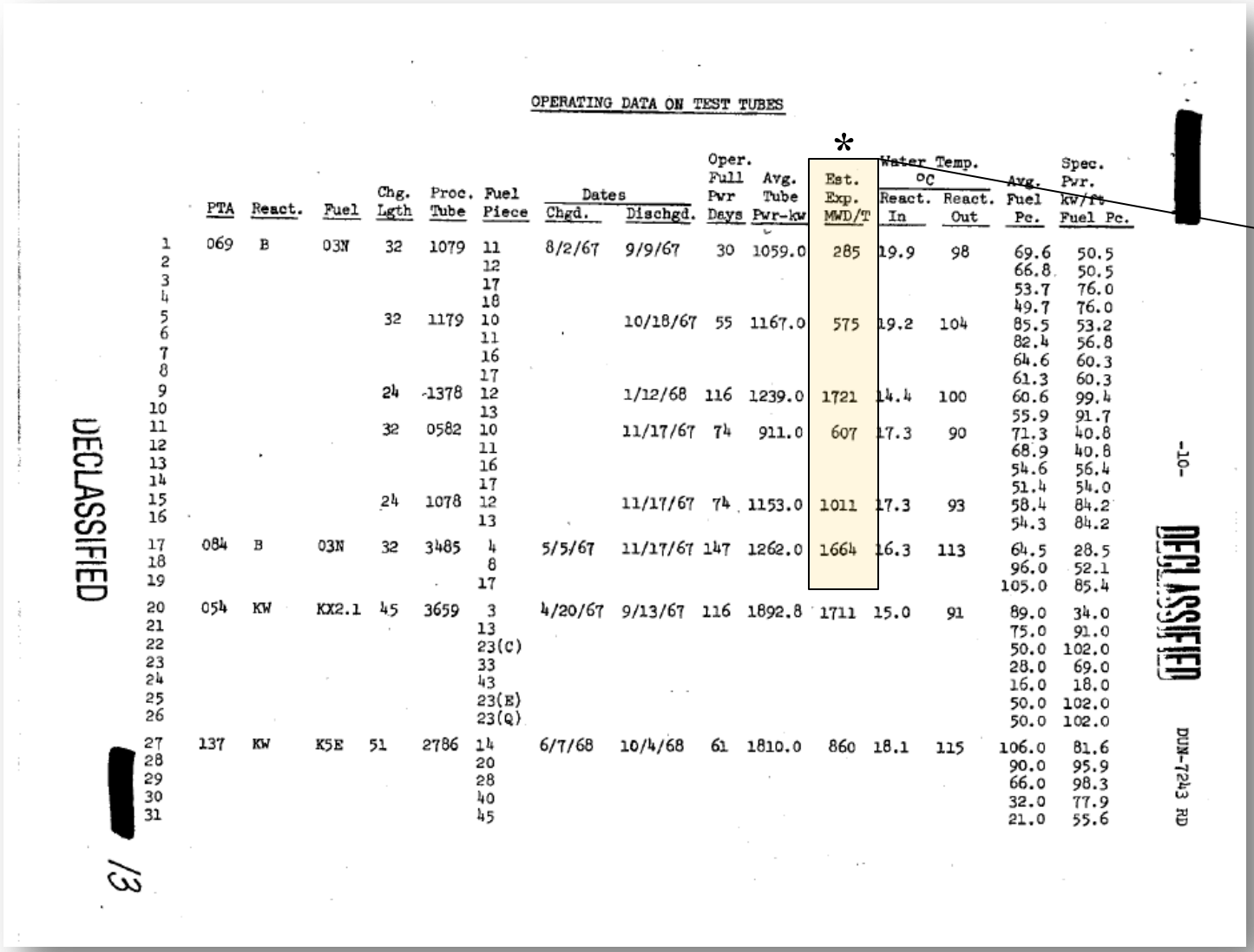
Hanford B Experiments (1967)

- **18 measurements from Hanford B selected from test programs PTA-069 and PTA-084 (^{237}Np production) for detailed evaluation using state-of-the-art methods and data**
- **Low exposure of natural uranium metal**
- **Destructive analysis of fuel**
  - Uranium isotopics (^{235}U < ±0.7% relative assay measurement uncertainty)
  - Plutonium isotopics (^{239}Pu < ±0.1%, ^{240}Pu < ±0.6%)
  - Plutonium generation (±3.5%)
  - Total Cs production (±3.0%)
  - Total Np production (±5.0%)
- **Different initial ^{236}U contents used to investigate ^{237}Np production**

PTA-069 and -084 ^{237}Np Experiments



OPERATING DATA ON TEST TUBES

	PTA	React.	Fuel	Chg. Lgth	Proc. Tube	Fuel Piece	Dates Chgd.	Dates Dischgd.	Oper. Full Pwr Days	Avg. Tube Pwr-kw	* Est. Exp. MWD/T	Water Temp. °C React. In	Water Temp. °C React. Out	Avg. Fuel Pc.	Spec. Pwr. kw/ft Fuel Pc.
1	069	B	03N	32	1079	11	8/2/67	9/9/67	30	1059.0	285	19.9	98	69.6	50.5
2						12								66.8	50.5
3						17								53.7	76.0
4						18								49.7	76.0
5				32	1179	10		10/18/67	55	1167.0	575	19.2	104	85.5	53.2
6						11								82.4	56.8
7						16								64.6	60.3
8						17								61.3	60.3
9				24	-1378	12		1/12/68	116	1239.0	1721	14.4	100	60.6	99.4
10						13								55.9	91.7
11				32	0582	10		11/17/67	74	911.0	607	17.3	90	71.3	40.8
12						11								68.9	40.8
13						16								54.6	56.4
14						17								51.4	54.0
15				24	1078	12		11/17/67	74	1153.0	1011	17.3	93	58.4	84.2
16						13								54.3	84.2
17	084	B	03N	32	3485	4	5/5/67	11/17/67	147	1262.0	1664	16.3	113	64.5	28.5
18						8								96.0	52.1
19						17								105.0	85.4
20	054	KW	KX2.1	45	3659	3	4/20/67	9/13/67	116	1892.8	1711	15.0	91	89.0	34.0
21						13								75.0	91.0
22						23(C)								50.0	102.0
23						33								28.0	69.0
24						43								16.0	18.0
25						23(E)								50.0	102.0
26						23(Q)								50.0	102.0
27	137	KW	K5E	51	2786	14	6/7/68	10/4/68	61	1810.0	860	18.1	115	106.0	81.6
28						20								90.0	95.9
29						28								66.0	98.3
30						40								32.0	77.9
31						45								21.0	55.6

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*
Burnup from 285 to 1721 MWd/ton U

Exposures from 30 to 147 days

^{240}Pu isotopic contents range from 3% to 17% of total Pu

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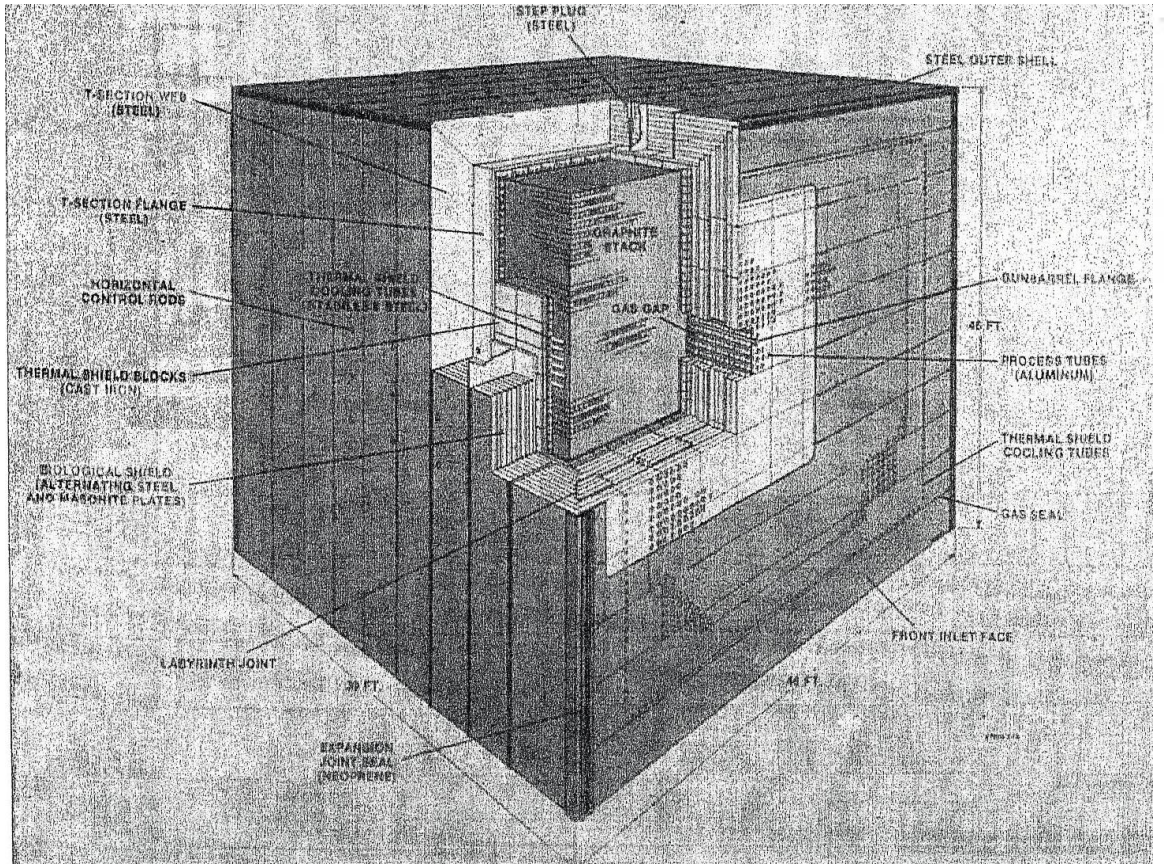
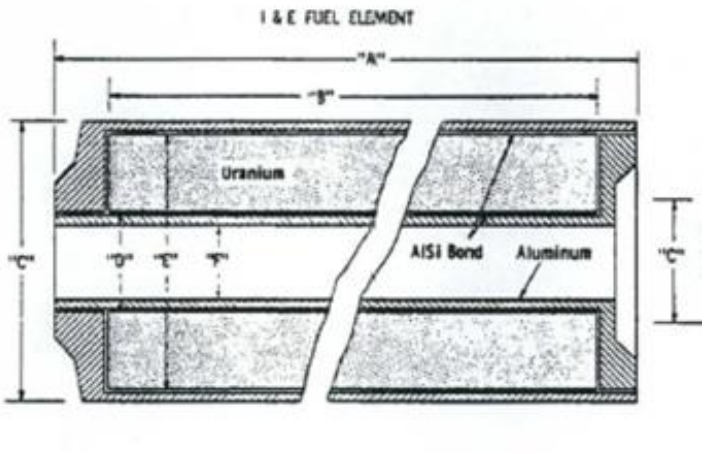
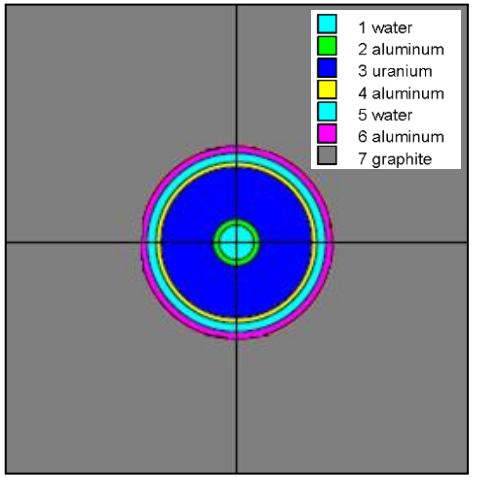
Hanford Reactor Model





Single process channel model



Hanford B reactor lattice (not to scale)

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Hanford B Reactor

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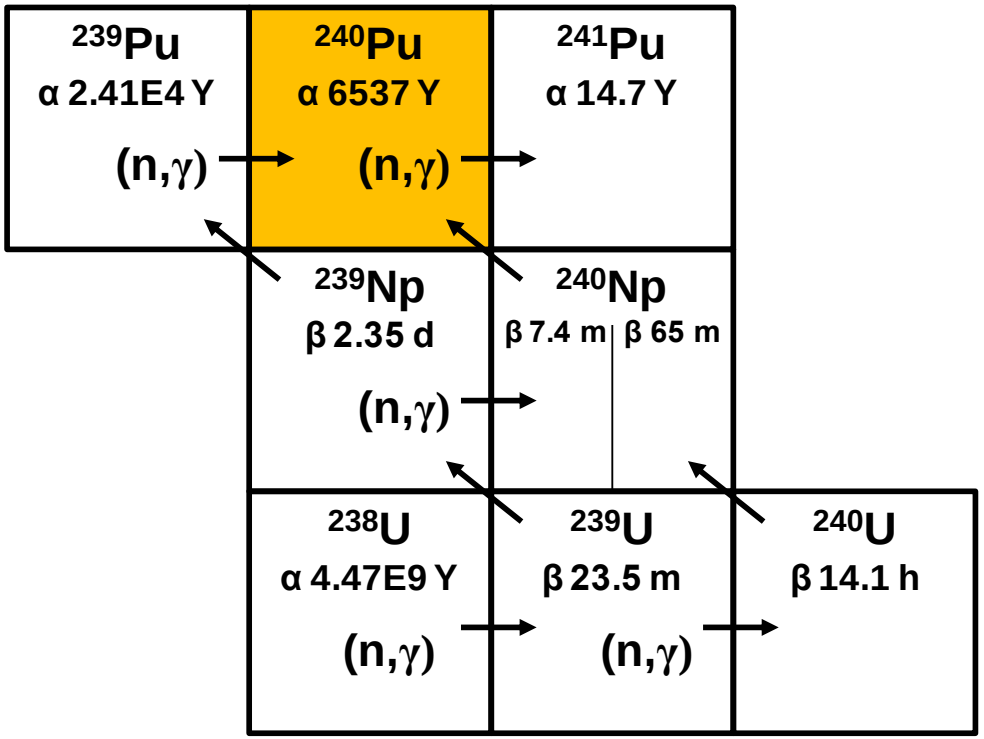
^{240}Pu Production Paths

- ^{239}U (n,γ) minor
- ^{239}Np (n,γ) very important at low exposures *
- ^{239}Pu (n,γ) dominant production route and higher burnup



*J. HALPERIN et al.
An Effective Capture Cross Section of Np^{239} for Thermal Reactor Neutrons, *Nuclear Science and Engineering* **1**, 108-111 (1956)

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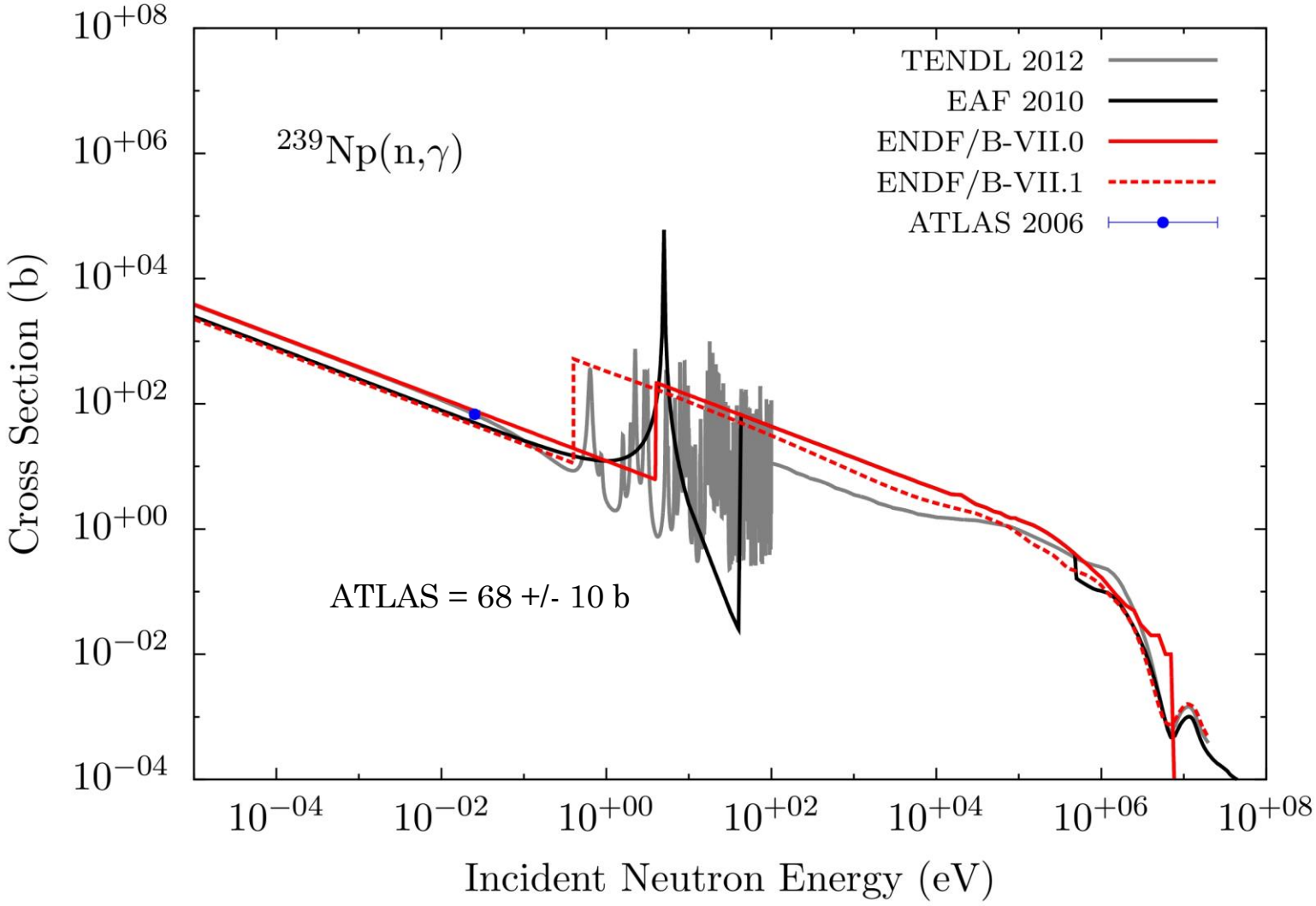
Computation Analysis

- Calculations performed using the SCALE nuclear systems modeling and simulation code suite
- Data processing using continuous-energy treatment in CENTRM module
- Cross sections applied in the ORIGEN code use to simulate results for 18 measured samples
- Analyses performed using ENDF/B-VII.0, -VII.1, JEFF-3.0/A, and TENDL cross sections

Current Evaluated Data for ^{239}Np (n,γ)



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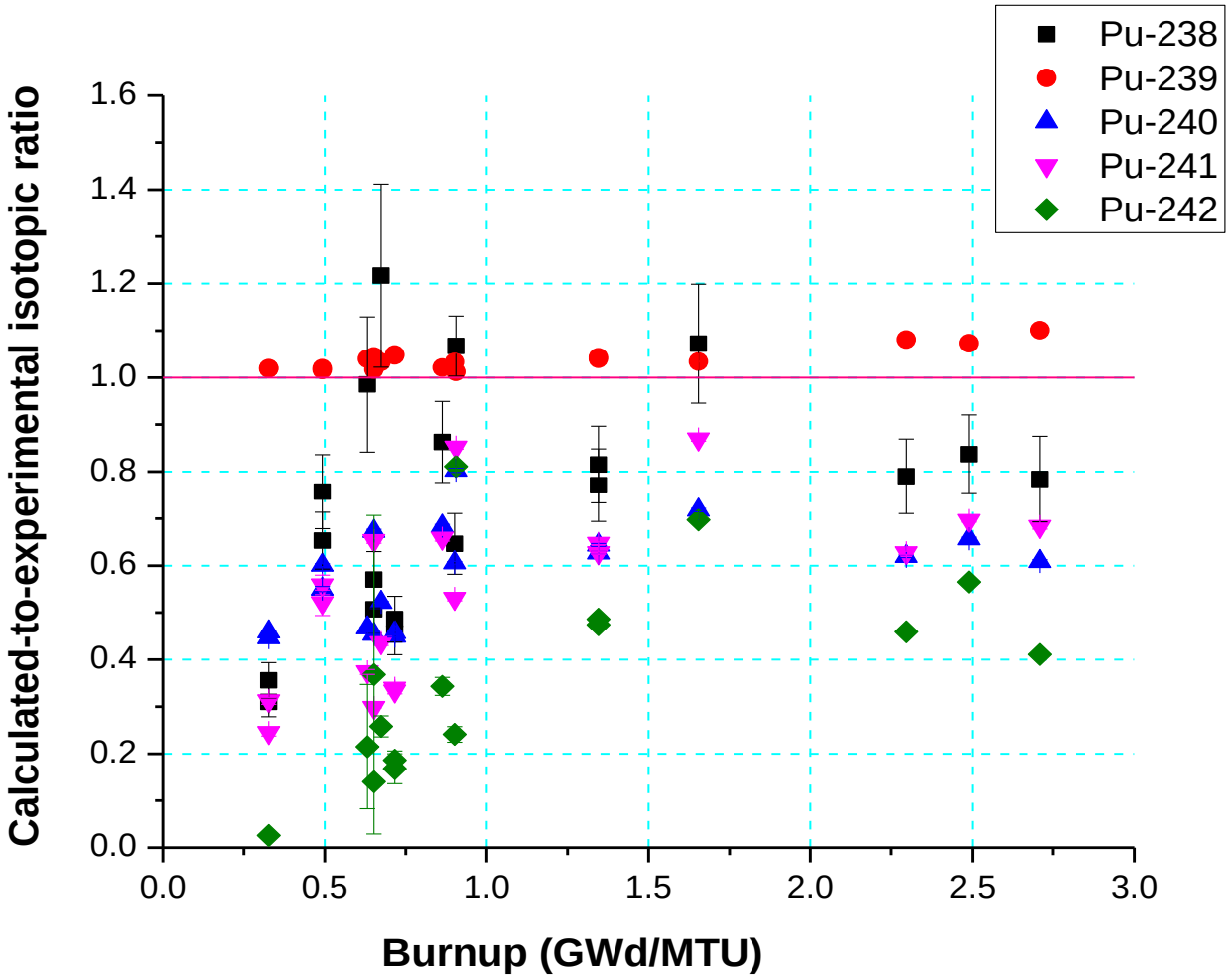
^{239}Np (n,γ) Cross Sections

Library	Capture cross section (b) *
ENDF/B-VII.0	28.0
ENDF/B-VII.1	35.2
JEFF -3.0/A (EAF)	49.0
TENDL	28.7

* Effective 1-group value for Hanford B reactor

ENDF/B-VII.0 Results (Pu isotopics)



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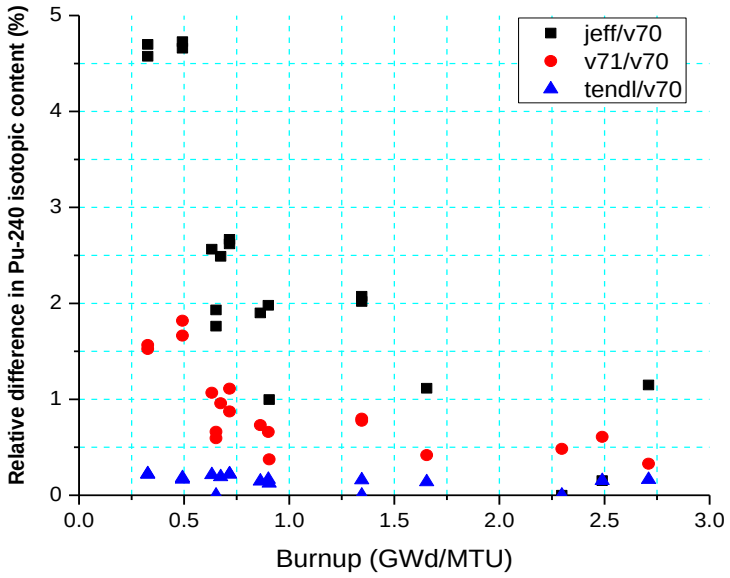
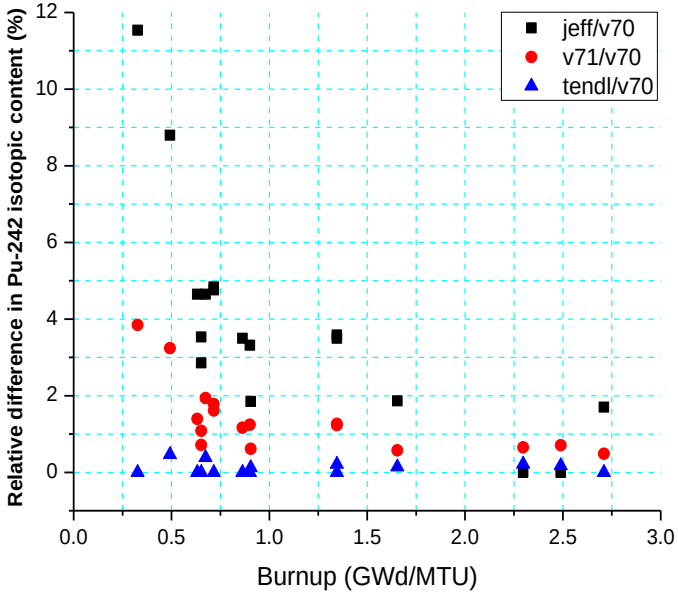
Effect of Nuclear Data Library

Relative change in predicted isotope content

^{240}Pu



^{242}Pu



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Conclusions

- Current evaluated nuclear data libraries appear to dramatically underestimate ^{240}Pu production in very low burnup uranium
- Measurements (< 1% uncertainty) indicate a systematic underestimate of production likely via the ^{239}Np capture route
- Different evaluations do not reconcile the errors
- Many measurements used in current cross section evaluation are from 1950s; analysis of measurements relied on low accuracy nuclear data
- There is a need to reanalyze earlier measurements using present day nuclear data and modern modeling and simulation tools
- Hanford experiments (1967) document important high quality mass spectrometry data that are currently being evaluated at ORNL to support improved nuclear data for low exposure fuels for safeguards/criticality safety applications

Acknowledgements

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