

Isotopic Validation of BWR Depletion Calculations

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Project Goals

- Establish uncertainty margins for BWR burnup credit due to predicted
 - Quantify uncertainties in predicted isotope concentrations by the lattice physics depletion code (validation).

Select a set of representative RCA (radiochemical assay) measurements in the application range

Model operating history of the fuel sample in the reactor

Compare measured and predicted isotope concentrations

Calculate k_{eff} differences for application of interest

Experimental Assay Data

- Nuclide compositions from 77 samples from assemblies irradiated in eight different reactors.
- Assembly designs include 6x6, 8x8, 9x9, and 10x10 configurations in SVEA (water cross), ATRIUM(water box) and GE (water rod) lattices.
 - Enrichments from 2.1 wt% to 4.94 wt% ^{235}U and up to 5 wt% Gd_2O_3 .
 - Burnups from 7 to 68 GWd/MTU
 - Average void fractions from 0% to 74%
- Measured assay data are obtained from:
 - OECD/NEA SFCOMPO Database (Dodewaard, Forsmark 3, Fukushima Daini 1 and 2)
 - Spanish Nuclear Safety Council (Forsmark 3)
 - MALIBU International Program (Leibstadt 3)
 - US DOE Yucca Mountain Project (Limerick 1)

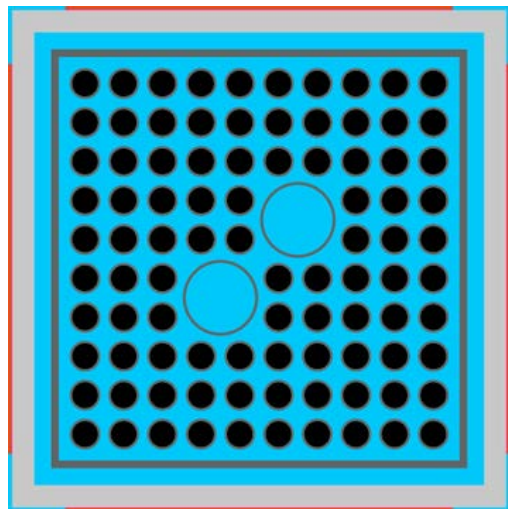
Experimental Assay Data

Reactor and Unit	Country	Assembly Design	Number of Samples	Enrichments (wt % ²³⁵ U)	Burnup (GWd/MTU)
Dodewaard	Belgium	6x6	1	4.94	55
Forsmark 3	Sweden	10x10 (SVEA-96)	1	3.97	61
Forsmark 3	Sweden	10x10 (GE14)	8	3.95	38–50
Fukushima Daini 1	Japan	9x9-9	13	2.1, 4.9, 3.0 (Gd)	35–68
Fukushima Daini 2	Japan	8x8-4	25	3.4, 4.5, 3.4 (Gd)	9–59
Fukushima Daini 2	Japan	8x8-2	18	3.9, 3.4 (Gd)	7–44
Leibstadt 3	Switzerland	10x10 (SVEA-96)	3	3.9	56–63
Limerick 1	United States	9x9 (GE11)	8	3.95, 3.6 (Gd)	37–65

- 80% of samples are from Fukushima Daini Units 1 and 2
- Forsmark 3 (GE14), Leibstadt 3 (SVEA-96) and Limerick 1 (GE11) are from proprietary experimental programs
- Several data sets analyzed in previous studies were rejected due to insufficient operating history data or for being unrepresentative of modern designs

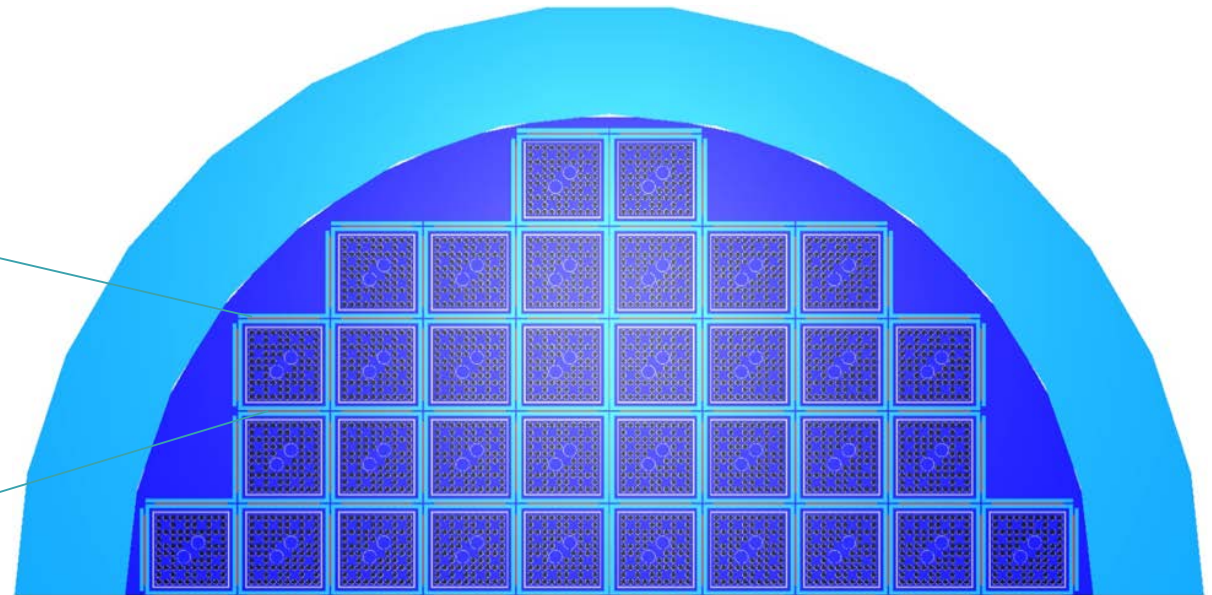
Application

GE14 fuel assembly



GBC-68 cask

- Stainless steel
- Water



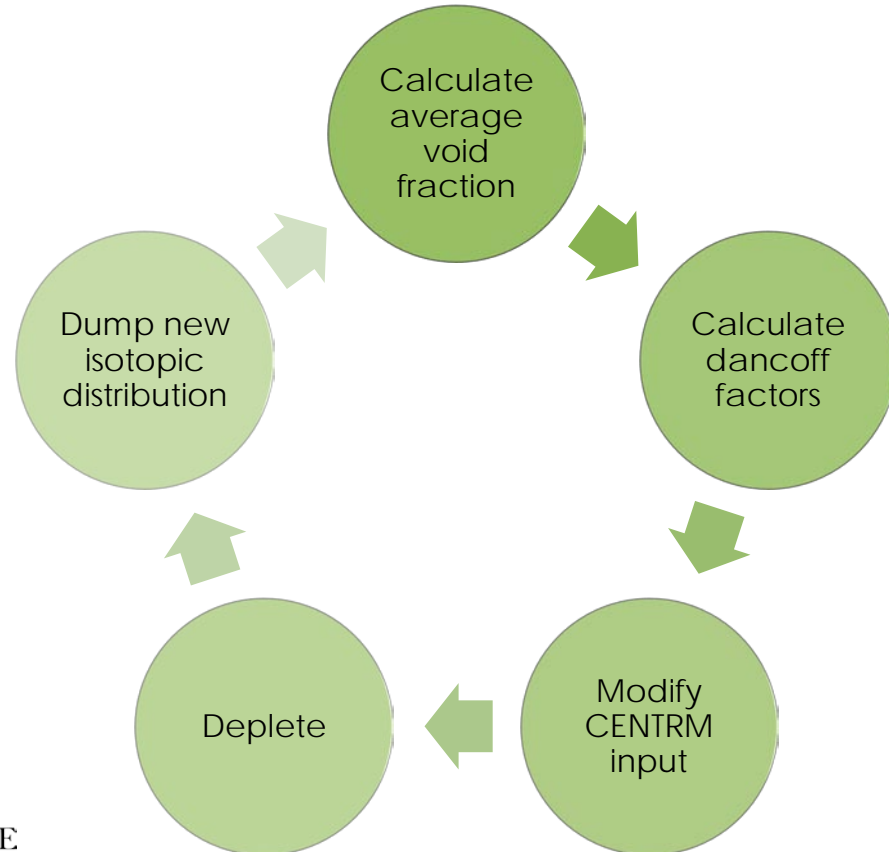
Predicting Nuclide Concentrations

- **TRITON/NEWT lattice physics code**

- General geometry
- Requires Dancoff factors calculated externally (MCDANCOFF)

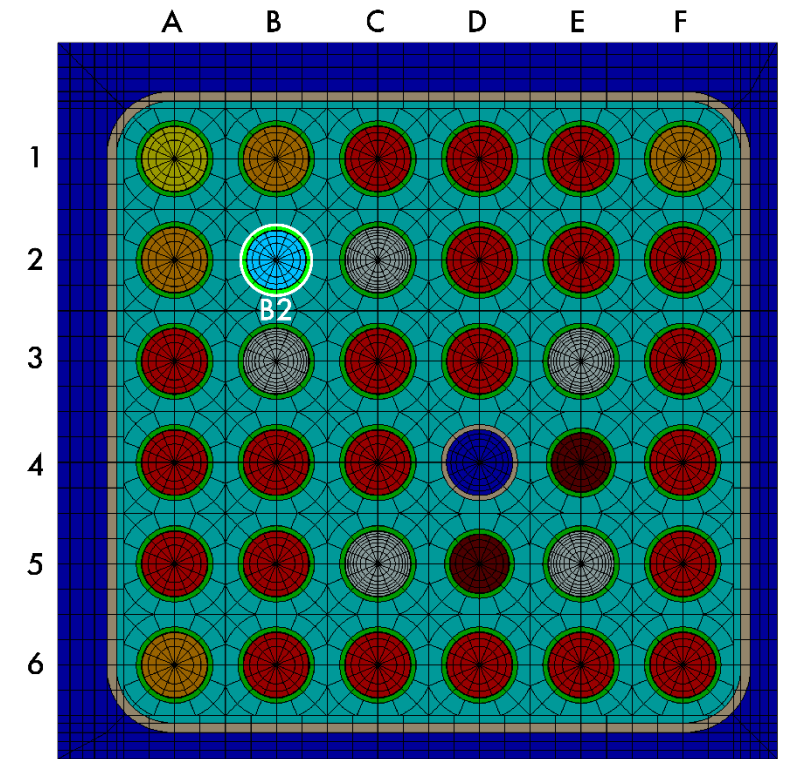
- **Polaris lattice physics code**

- Fast depletion calculations
- Simple input interface tailored for lattice physics calculations
- Effect of state changes (void fraction and fuel temperature) on cross section processing are included at every depletion steps implicitly



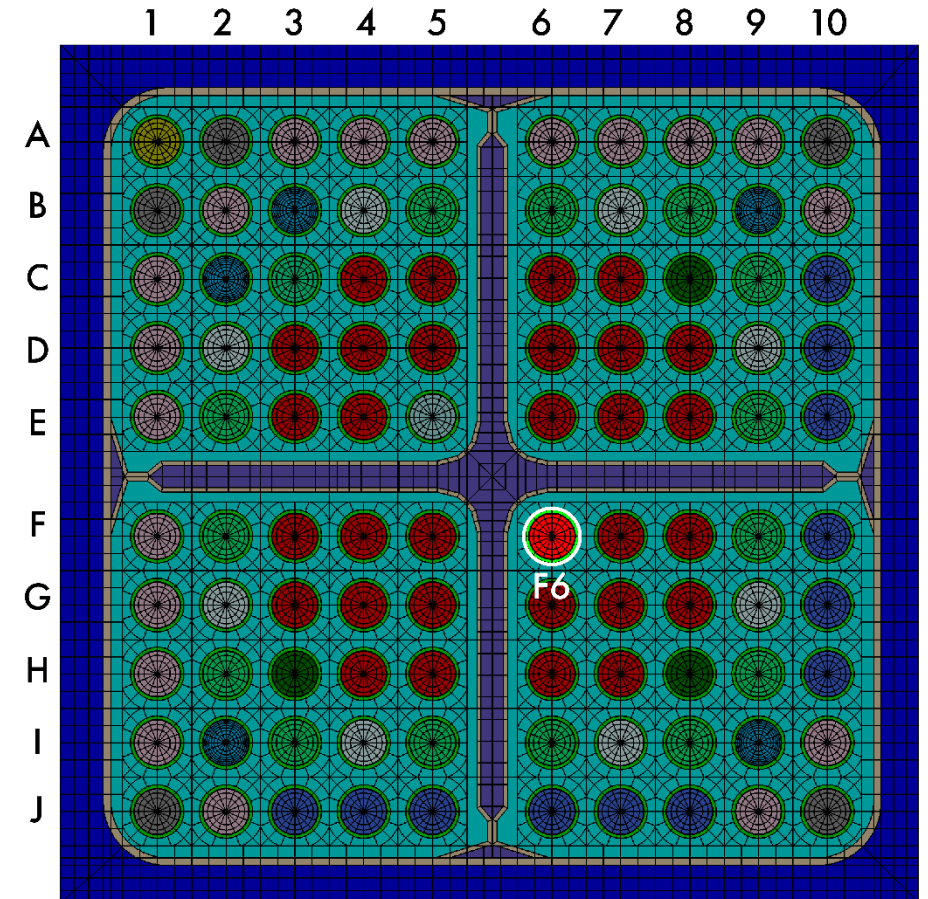
Dodewaard (1 Sample)

- JNF 6x6 design
- 5 cycles of operation
- One water rod, 5 Gd₂O₃ (2.7 wt%) loaded fuel rods
- Enrichments : 1.8 wt% to 4.94 wt% ²³⁵U
- Two MOX rods with 6.43 wt % total plutonium
- Average void fraction: 50%
- Assembly average burnup: 35.24 GWd/MTU
- Sample burnup: 55.5 GWd/MTU
- Sample burnup is matched to measured ¹⁴⁸Nd content
- There are two measurements for the same sample with large differences in actinide concentrations (e.g., 6 % difference in ²³⁵U)



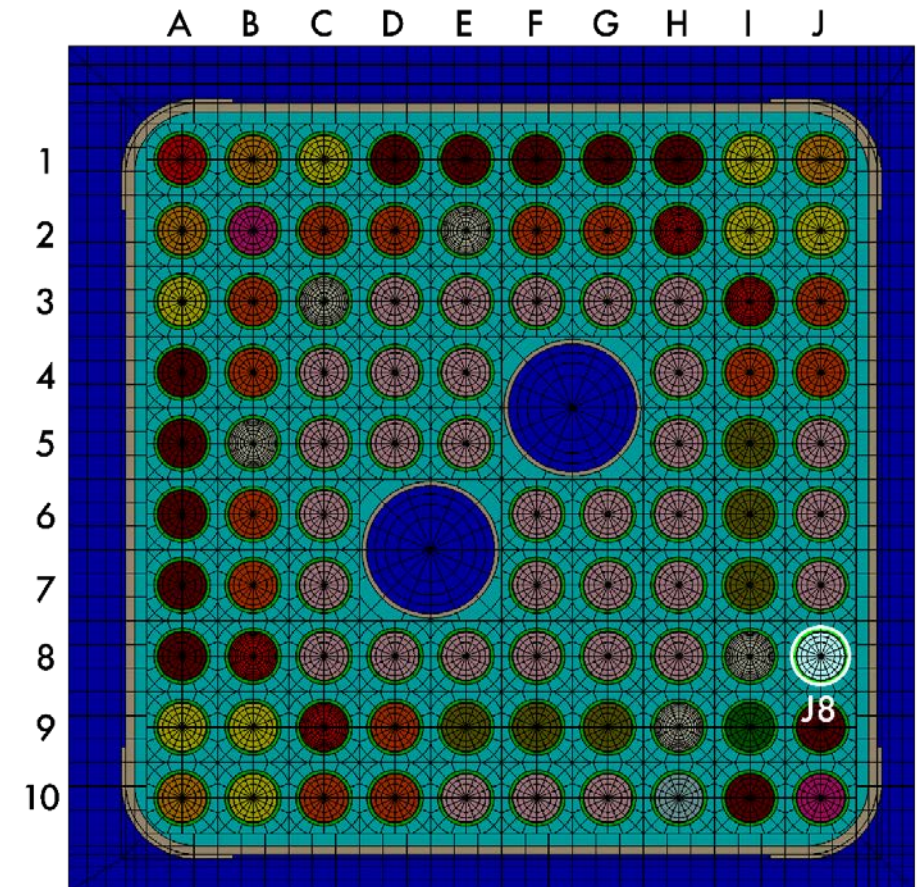
Forsmark 3 (1 Sample)

- SVEA-100 design
- 6 cycles of operation
- 5 Gd_2O_3 (3.15 wt%) loaded fuel rods
- Sample enrichment: 3.97 wt% ^{235}U
- Average void fraction: 58%
- Assembly average burnup: 50.67 GWd/MTU
- Sample burnup: 58 GWd/MTU
- Sample burnup is matched to ^{148}Nd content



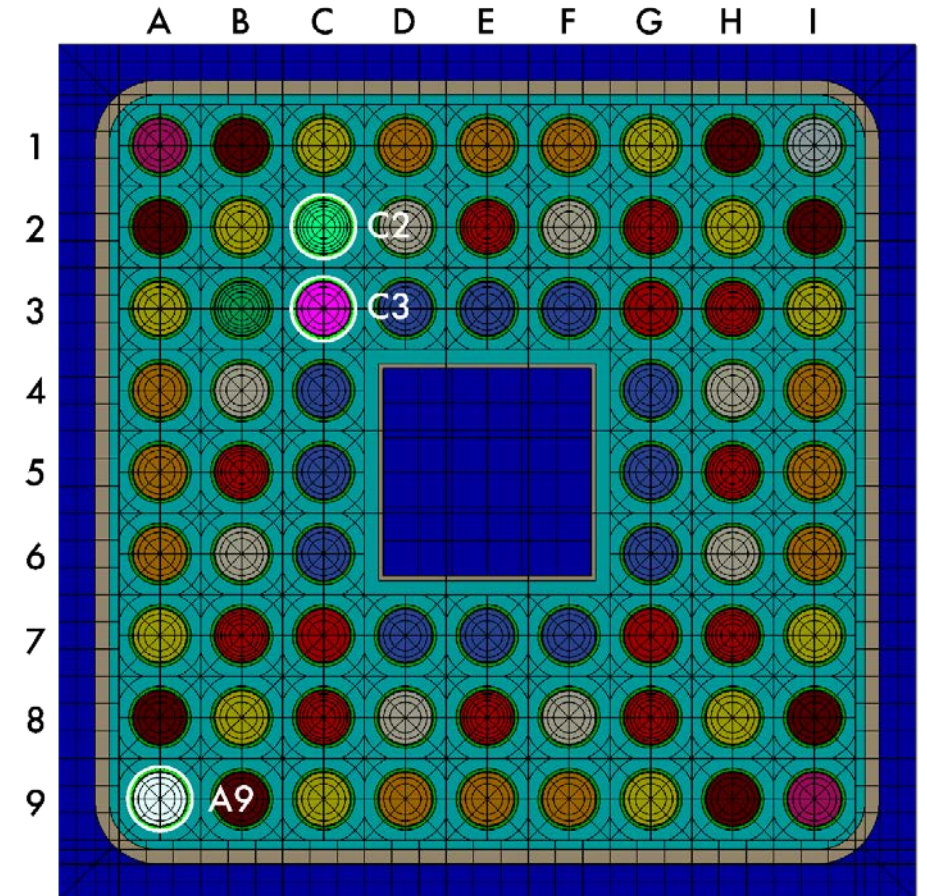
Forsmark 3 (8 Samples)

- GE 14x14 design
- 5 cycles of operation
- 9 Gd₂O₃ (4.0 wt%) loaded fuel rods
- 8 samples from the same fuel rod
- Sample enrichments: 3.95 wt% ²³⁵U
- Average void fraction: 2.2% to 67%
- Assembly average burnup: 47.27 GWd/MTU
- Sample burnups: 38.3 to 51.1 GWd/MTU
- Sample burnups are matched to measured ¹⁴⁸Nd content, except for one sample that is matched to gamma scan



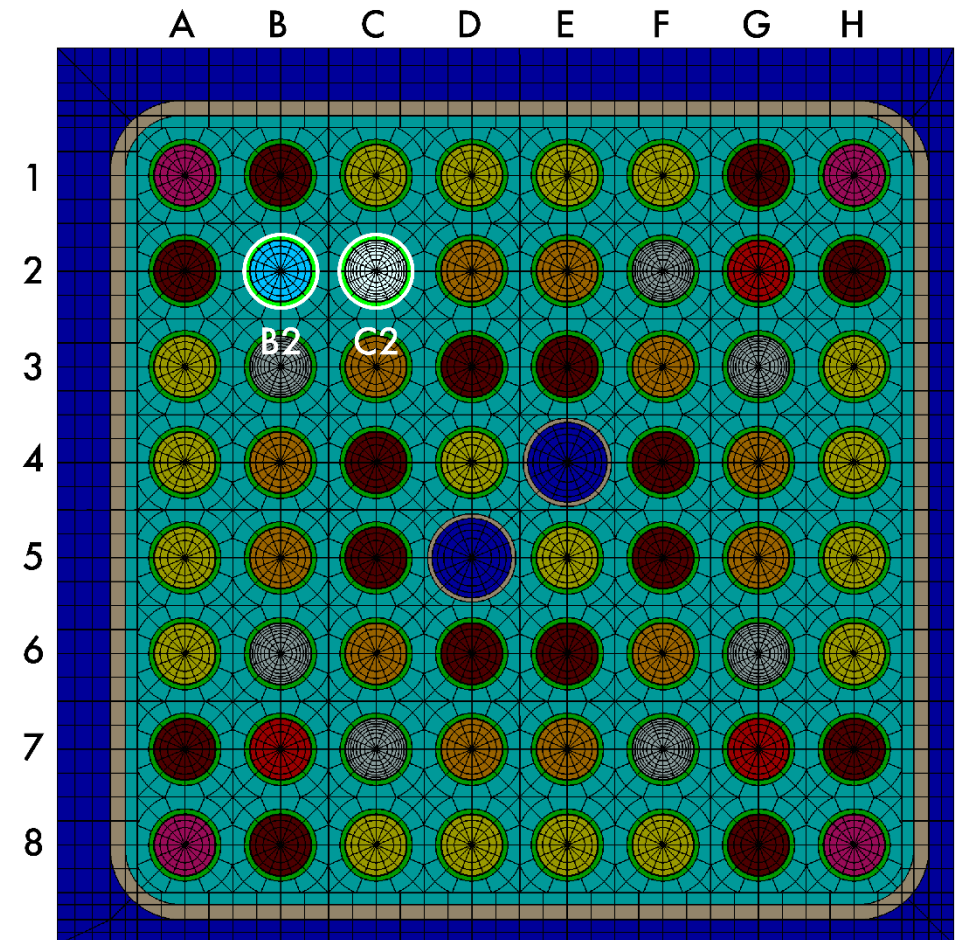
Fukushima Daini 1 (13 Samples)

- 9x9-9 design
- Two assemblies after 3 and 5 cycles of operation
- 8 Gd₂O₃ (5.0 wt%) loaded fuel rods
- 13 samples from three rods
 - 5 samples from Gd₂O₃ loaded rods
- Sample enrichments: 2.1 to 4.9 wt% ²³⁵U
- Void fractions: 11% to 74%
- Sample burnups: 35.6 to 68.4 GWd/MTU
- Sample burnups are matched to measured ¹⁴⁸Nd content



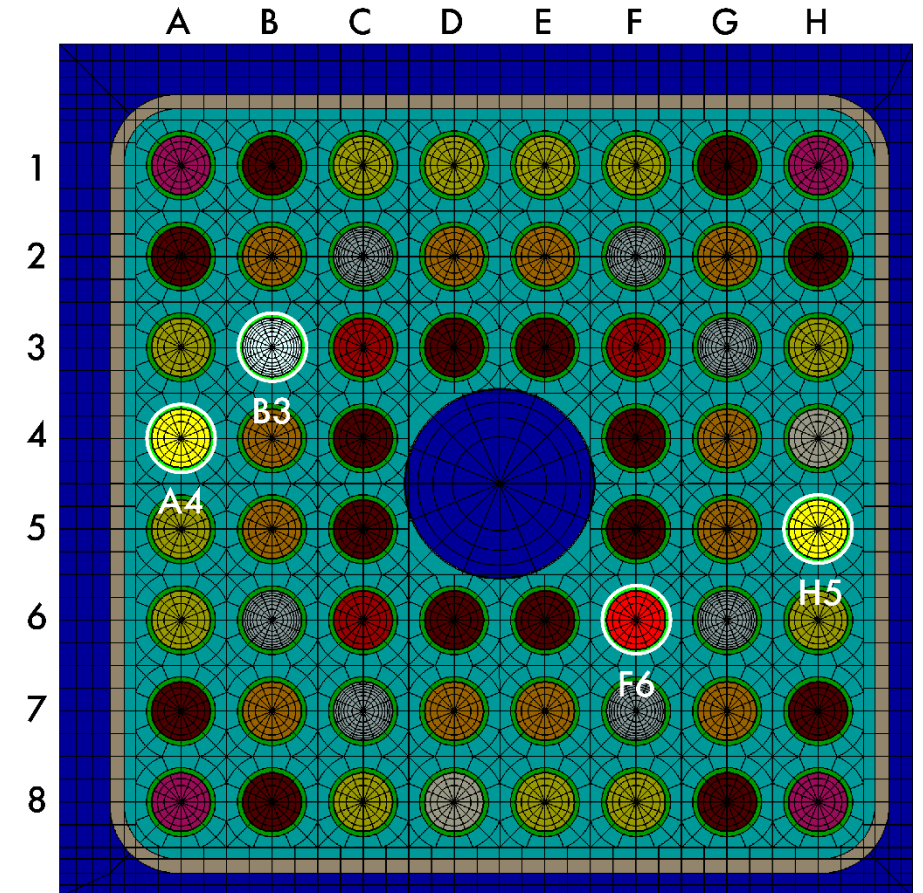
Fukushima Daini 2 (18 Samples)

- 8x8-2 design
- 3 cycles of operation
- 8 Gd₂O₃ (3.0-4.5 wt%) loaded fuel rods
- 18 samples from two fuel rods
 - 10 samples from 4.5 wt% Gd₂O₃ loaded rods
- Sample enrichments: 0.71 to 3.94 wt% ²³⁵U
- Void fractions: 0% to 74.3%
- Sample burnups: 4.2 to 44.0 GWd/MTU
- Sample burnups are matched to ¹⁴⁸Nd content
- Only reactor average nodal void fractions are available



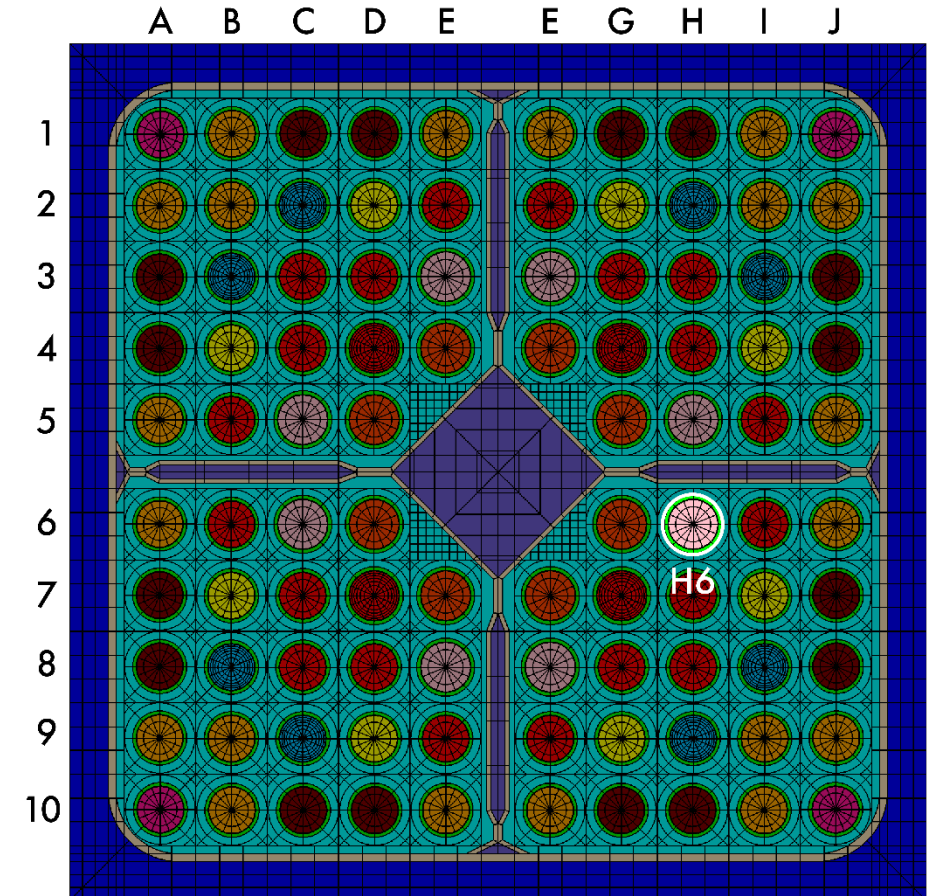
Fukushima Daini 2 (25 samples)

- 8x8-4 design
- 1, 2, 3 and 5 cycles of operation
- 25 samples from 11 fuel rods in 4 assemblies
 - 11 samples from 4.5 wt% Gd_2O_3 loaded rods
- 8 Gd_2O_3 (3.0-4.5 wt%) loaded rods
- Sample enrichments : 3.4 and 4.5 wt% ^{235}U
- Void fractions: 0% to 74.3%
- Sample burnups: 9.4, 59.1 GWd/MTU
- Sample burnups are matched to measured ^{148}Nd content



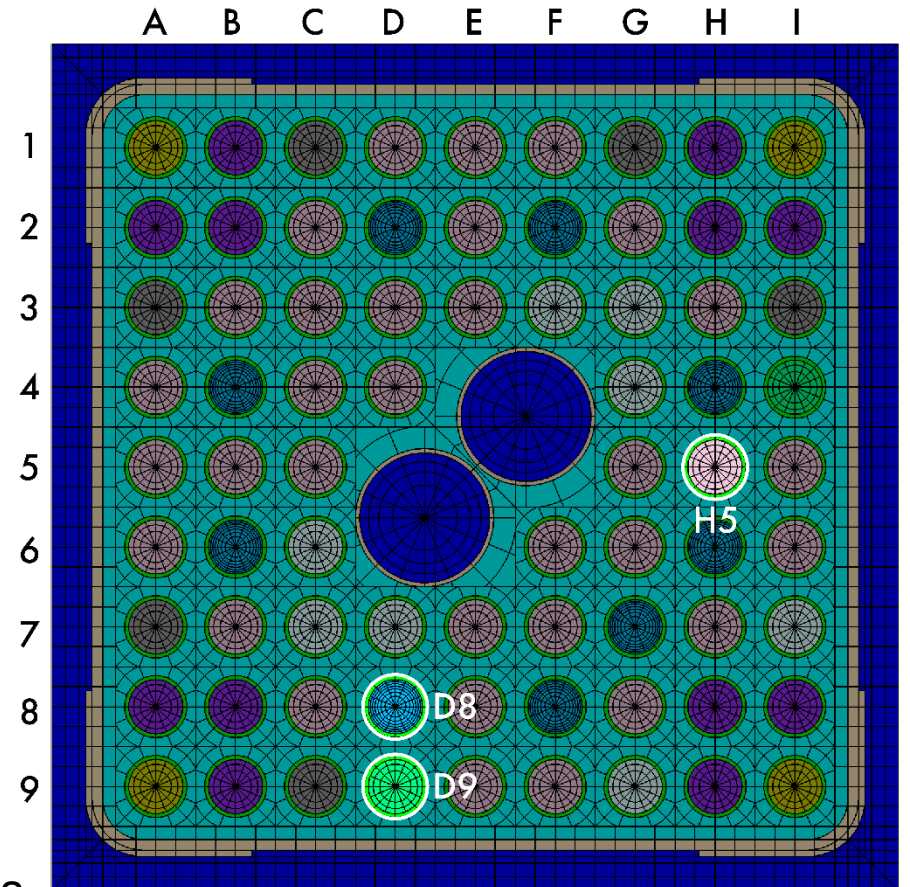
Leibstadt (3 Samples)

- SVEA-96 design
- 7 cycles of operation
- 3 samples from the same fuel rod
- 12 Gd₂O₃ (4.0 wt%) loaded fuel rods
- Sample enrichment: 3.9 wt% ²³⁵U
- Void fractions: 8.4, 51 and 70%
- Sample burnups: 58.4, 60.5 and 65.0 GWd/MTU
- Sample burnups are matched to measured ¹⁴⁸Nd content except for one sample matched to measured ¹⁴⁶⁺¹⁴⁵Nd and ¹³⁷Cs content



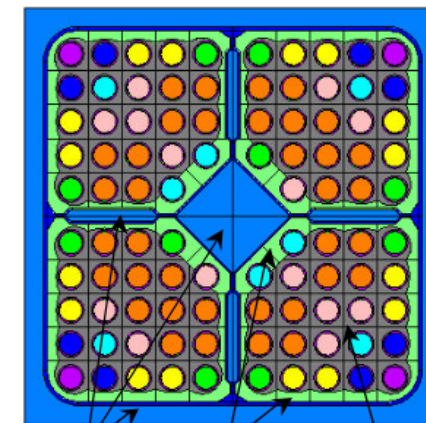
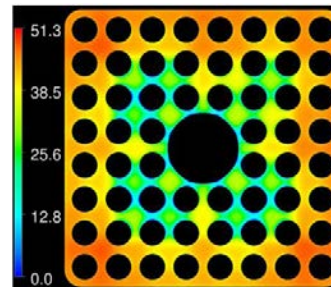
Limerick 1 (8 Samples)

- GE11 9x9 design
- 3 cycles of operation
- 9 Gd₂O₃ (5.0 wt%) loaded fuel rods
- 8 samples from three fuel rods
 - 2 samples from 4.5 wt% Gd₂O₃ loaded rods
- Sample enrichments: 3.9 wt% ²³⁵U
- Void fractions: 12 to 69%
- Sample burnups: 37 to 65.5 GWd/MTU
- Sample burnups are matched to declared burnup.
 - Declared burnups were calculated using ratios of major actinides to neodymium isotopes.
 - Burnups matched to measured ¹⁴⁸Nd result in inconsistent actinide predictions.

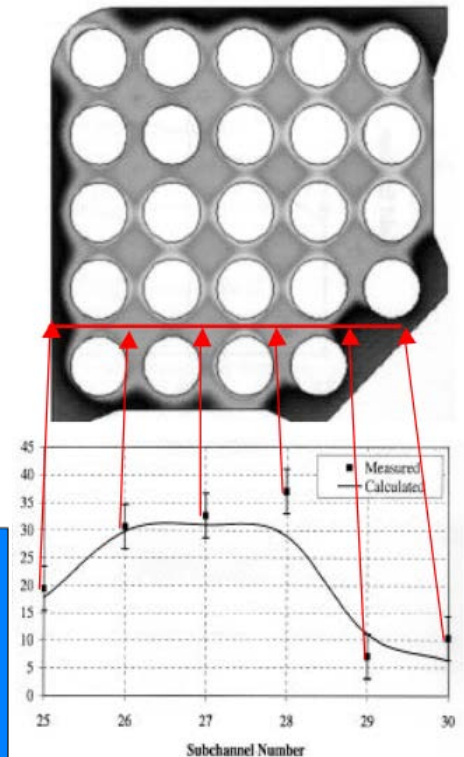


Modeling Challenges

- Void fraction history
 - Major source of uncertainty for modeling
 - Several measurements were rejected because of missing void fraction history
 - Only lattice average void fraction is provided
 - 5.3% and 6.3% uncertainty in lattice average void fraction by transient and sub-channel codes.
 - Void fraction distribution in regions near the channel, corner or water rods can be 25% less than the average void fraction.
 - Based on axial location of the sample, effect of void fraction uncertainty on k-effective and isotope distribution can be large.... more discussion on this later



0% void 4.8% void 35% void



"Effects Of Void Uncertainties On Pin Power Distributions And The Void Reactivity Coefficient For A 10x10 BWR Assembly" F. Jatuff et.al.

Modeling Challenges (cont.)

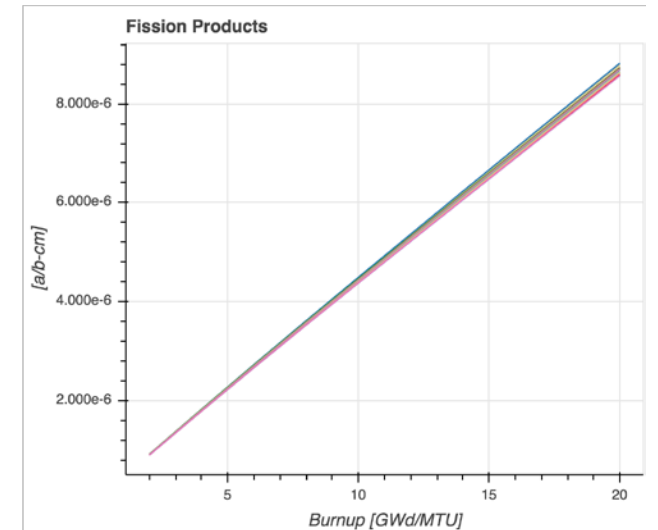
– Fuel Temperature History

- Rarely available. Although a detailed history is not required, a reasonable value should be used based on fuel rod diameter.
- Effective fuel temperature uncertainty of 100 K causes 200 pcm difference in the calculated k_{eff} .

– Burnup

- Generally estimated using measured ^{148}Nd concentrations. Alternative burnup indicator i.e., ^{137}Cs , $^{145}\text{Nd} + ^{146}\text{Nd}$ are also used
- What if burnups calculated from these burnup indicators do not match?

Sample	Burnup [GWd/MTU] based on						Gamma S.
	$^{146}\text{Nd}/^{238}\text{U}$	$^{148}\text{Nd}/^{238}\text{U}$	$^{150}\text{Nd}/^{238}\text{U}$	^{235}U	^{239}Pu		
Forsmark-4	51.9	65.0	55.9	50.6	48.2	53.9	
Uncertainty	0.9	4.3	1.8	0.3	0.3		



^{148}Nd Concentration vs Burnup

Polaris results actinides (C/M-1)

Data	Number of Measurements	Mean	Standard Deviation	Median	Minimum	Maximum	1st Quartile (Q1)	3rd Quartile (Q3)	Percentile P10 ^a	Percentile P90 ^a
U-234	76	6.8%	13.4%	5.5%	-37.0%	66.6%	1.4%	9.9%	-5.4%	20.6%
U-235	76	4.3%	11.2%	2.8%	-15.1%	36.5%	-2.8%	9.1%	-8.1%	22.0%
U-236	76	1.6%	4.8%	1.2%	-6.0%	15.7%	-1.5%	3.2%	-3.4%	7.7%
U-238	76	-0.1%	0.3%	-0.1%	-0.8%	0.5%	-0.2%	0.1%	-0.6%	0.2%
Pu-238	76	9.5%	21.1%	6.7%	-38.8%	93.6%	-1.9%	20.1%	-18.1%	33.2%
Pu-239	76	-0.9%	8.7%	-1.0%	-22.8%	22.7%	-6.6%	4.4%	-11.6%	10.2%
Pu-240	76	-3.1%	8.4%	-2.4%	-28.3%	31.4%	-6.9%	-0.5%	-12.6%	6.5%
Pu-241	76	-3.3%	11.6%	-2.4%	-34.5%	40.1%	-8.8%	2.9%	-17.6%	9.2%
Pu-242	76	1.2%	17.2%	1.2%	-42.6%	87.9%	-4.5%	9.6%	-19.8%	15.1%
Am-241	62	2.8%	17.4%	3.6%	-50.3%	69.1%	-5.0%	10.5%	-15.2%	15.6%
Am-243	62	1.3%	33.5%	-7.5%	-44.5%	122.8%	-16.6%	4.9%	-26.7%	49.9%
Np-237	29	-2.4%	11.9%	-5.7%	-19.8%	46.4%	-7.7%	-1.1%	-11.3%	6.1%

*P10 and P90 and 10% and 90% percentiles of the distribution of the C/E-1 deviations

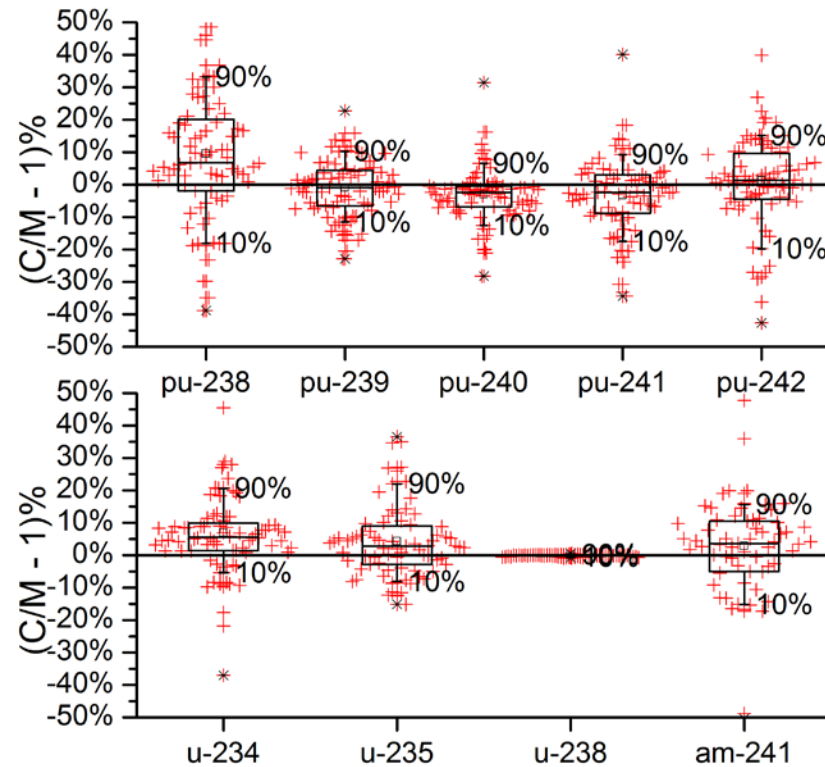
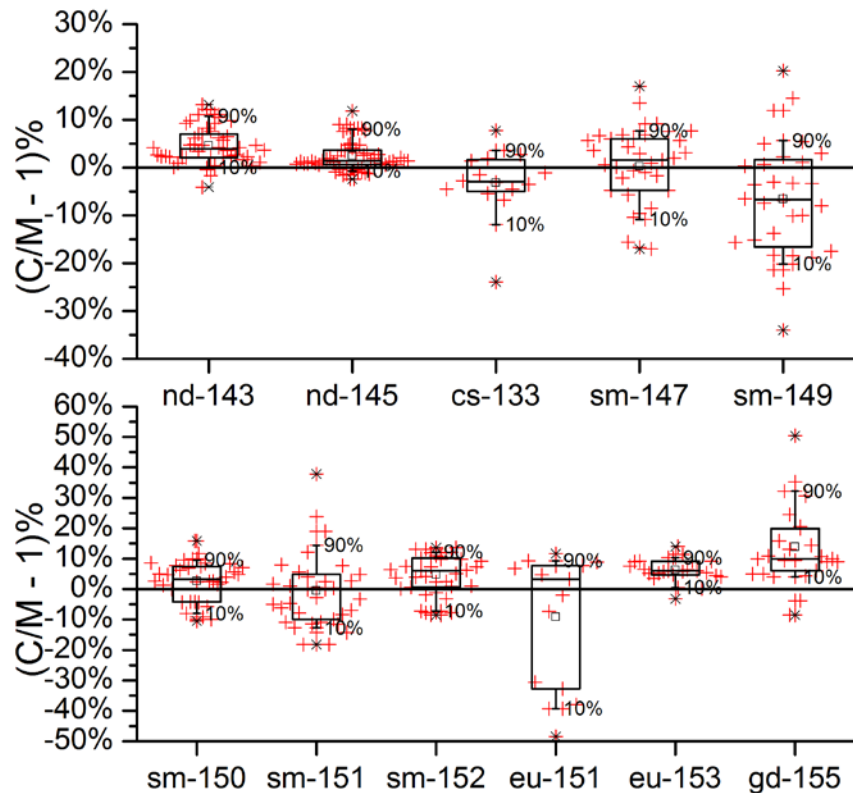
Polaris results fission products

Data	Number of Measurements	Mean	Standard Deviation	Median	Minimum	Maximum	1st Quartile (Q1)	3rd Quartile (Q3)	Percentile P10*	Percentile P90*
Nd-143	50	4.5%	4.0%	3.9%	-4.1%	13.1%	2.1%	7.0%	0.0%	10.8%
Nd-145	50	2.5%	3.2%	1.4%	-2.5%	11.8%	0.7%	3.6%	-0.8%	8.0%
Mo-95	23	2.0%	7.7%	-0.4%	-11.5%	17.6%	-3.1%	5.0%	-4.8%	14.5%
Tc-99	16	25.5%	15.5%	23.3%	-4.3%	49.5%	14.6%	38.7%	6.0%	45.3%
Ru-101	14	5.5%	13.4%	4.2%	-4.7%	48.8%	-2.2%	7.4%	-3.3%	9.5%
Ag-109	15	31.0%	38.1%	20.2%	-17.8%	147.2%	12.7%	46.4%	-4.5%	54.5%
Cs-133	16	-3.2%	7.2%	-2.9%	-24.0%	7.7%	-5.0%	1.7%	-11.9%	3.6%
Sm-147	35	0.2%	8.2%	1.6%	-17.0%	17.0%	-4.8%	6.0%	-10.8%	7.7%
Sm-149	32	-6.6%	12.2%	-6.7%	-34.0%	20.2%	-16.6%	1.6%	-20.2%	5.6%
Sm-150	34	2.6%	6.6%	3.3%	-10.4%	15.8%	-4.2%	7.5%	-8.1%	9.6%
Sm-151	35	-0.5%	11.9%	-0.2%	-18.2%	37.9%	-10.0%	4.9%	-12.7%	14.4%
Sm-152	35	4.7%	6.5%	6.0%	-8.5%	13.6%	0.7%	10.2%	-7.1%	12.2%
Eu-151	15	-9.2%	21.8%	3.2%	-48.4%	11.7%	-32.7%	7.7%	-39.3%	9.3%
Eu-153	25	6.3%	3.7%	6.0%	-3.2%	14.0%	4.7%	9.2%	0.6%	10.3%
Gd-155	25	13.9%	12.8%	10.0%	-8.5%	6.0%	10.0%	19.9%	50.4%	4.0%
Rh-103	15	5.2%	9.0%	2.5%	-6.2%	31.1%	1.1%	5.0%	-0.3%	20.2%

*P10 and P90 and 10% and 90% percentiles of the distribution of the C/E-1 deviations

Comparison of Measured and Calculated Nuclide Concentrations

- Only used to provide missing (not measured) isotopes and associated uncertainties



Isotope	Number of Measurements
²³⁴ U	77
²³⁵ U	77
²³⁸ U	77
²³⁸ Pu	77
²³⁹ Pu	77
²⁴⁰ Pu	77
²⁴¹ Pu	77
²⁴² Pu	77
²⁴¹ Am	63
²³⁶ U	37
²³⁷ Np	29
²⁴³ Am	37
⁹⁵ Mo	20
⁹⁹ Tc	12
¹⁰¹ Ru	12
¹⁰³ Rh	12
¹⁰⁹ Ag	12
¹³³ Cs	12
¹⁴⁷ Sm	37
¹⁴⁹ Sm	37
¹⁵⁰ Sm	37
¹⁵¹ Sm	37
¹⁵² Sm	37
¹⁴³ Nd	51
¹⁴⁵ Nd	51
¹⁵¹ Eu	12
¹⁵³ Eu	20
¹⁵⁵ Gd	20

Sources of Uncertainties in k_{eff}

- Measurements

- Uncertainties in the measured nuclide concentrations are introduced due to the complexity of radiochemical analysis
- Biases of 500 pcm were observed when measurements of the same sample made by two different laboratories were used in k_{eff} calculations
- Measurement uncertainties propagate to sample burnup estimates. Burnup uncertainties up to 6.5% were reported, resulting in maximum 2000 pcm uncertainty in calculated k_{eff}

- Calculations

- The input data associated with the operating history (void fraction, fuel temperature) are estimated by the operator and can have large uncertainties
- 5–6% uncertainty in average axial segment void fraction and larger uncertainties (up to 25%) for local void conditions radially within the assembly can result in more than 1500 pcm uncertainty in k_{eff} for some samples

Conclusion

- The mean values of C/M ratios for major actinides and fission products are comparable to previous validation work for BWR and PWR samples. However, large standard deviations are observed for some isotopes (^{238}Pu , ^{241}Am , ^{151}Eu , ^{149}Sm).
- A small number of samples for fission products (e.g., only 12 samples for ^{151}Eu) may contribute to large standard deviations.
- In general large differences in C/M ratios are observed in samples with large measurement and/or modeling uncertainties.
 - Uncertainties in complex radiochemical analysis techniques for some isotopes.
 - Uncertainties in void fraction distribution and sample burnup (the largest contributor).