

# Decay heat calculations

## A study of their validation and accuracy.

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# The UK National Nuclear Laboratory

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The NNL ([www.nnl.co.uk](http://www.nnl.co.uk)) was setup in April 2009 from R&D departments in subsidiaries of the former British Nuclear Fuels (BNFL) owned by the UK Government.

The NNL purposes include:

- Identifying and preserving key nuclear scientific and technical skills and facilities that cannot be reliably supplied by the marketplace.
- Providing independent and trusted technical advice to the UK Government and its agencies.

It consists of 687 staff, 614 of which are professionals working in 29 key skill areas.

# Why is decay heat important

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Important for engineering safety because of temperature induced damage to fuel and surroundings, for example

- Reactors

- Will recoverable trips lead to fuel damage
- Will severe accidents lead to release of radionuclides

- Fuel cycles / direct disposal

Storage and transport of spent fuel

Chemical separation processes

Geological disposal

- Handling difficulties due to damaged fuel/waste form
- Equipment failure due to boiling/melting
- Breach of containment releasing radionuclides



# Decay heat

Decay Heat can be calculated from the activities and energy release per decay of the nuclides present.

$$H = \sum N_i \lambda_i E_i$$

In the UK, FISPIN is used to calculate spent fuel compositions, using the standard equations:

$$\begin{aligned} \frac{dN_i}{dt} = & -\lambda_i N_i + \sum_j \lambda_j N_j B_{j,i} \\ & + \sum_k N_k \sigma_{f,k} \phi Y_{k,i} - \sum_l N_i \sigma_{i,l} \phi + \sum_m N_m \sigma_{m,i} \phi \end{aligned}$$

# Decay Heat Validation

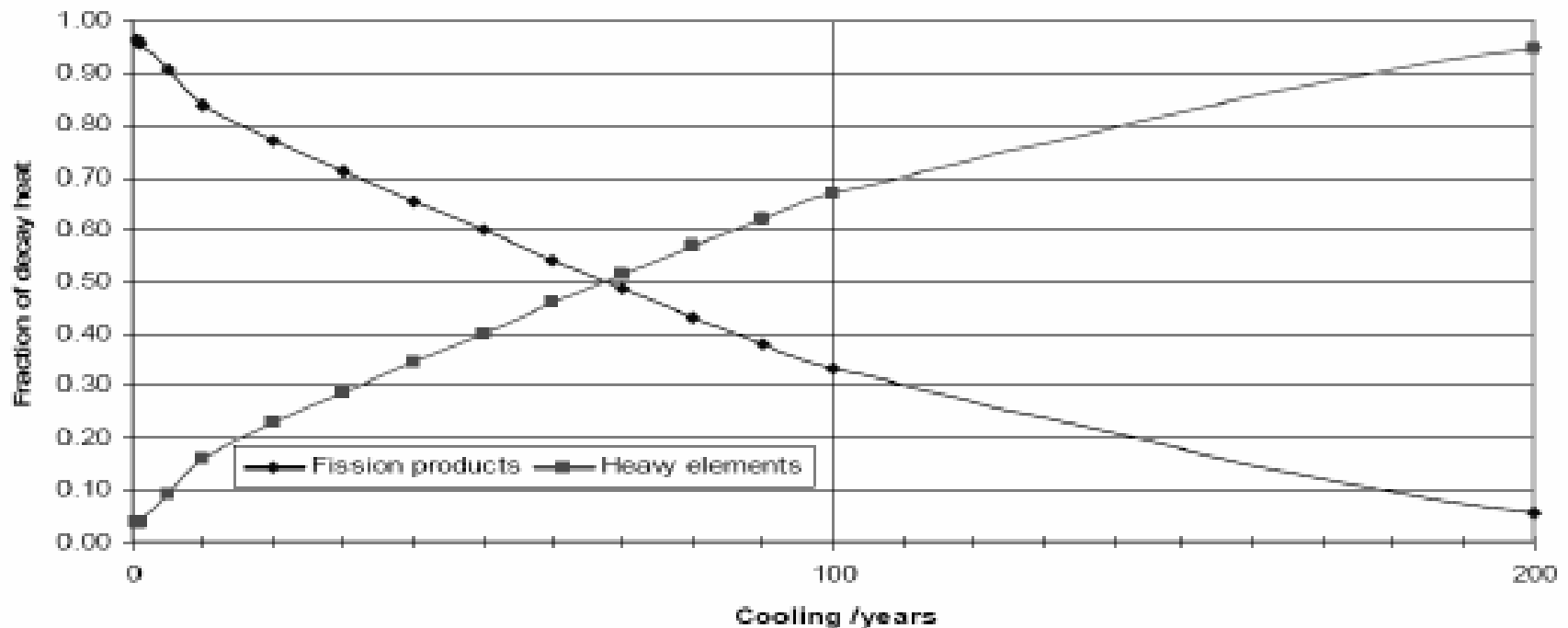
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How accurately can decay heat be estimated?

- Short term (seconds to days)
  - Reactors
  - Many nuclides important
- Mid term (years to decades)
  - Storage, transport, chemical processing
  - Small number of nuclides important
- Long term (centuries to millions of years)
  - Geological disposal
  - Few nuclides important

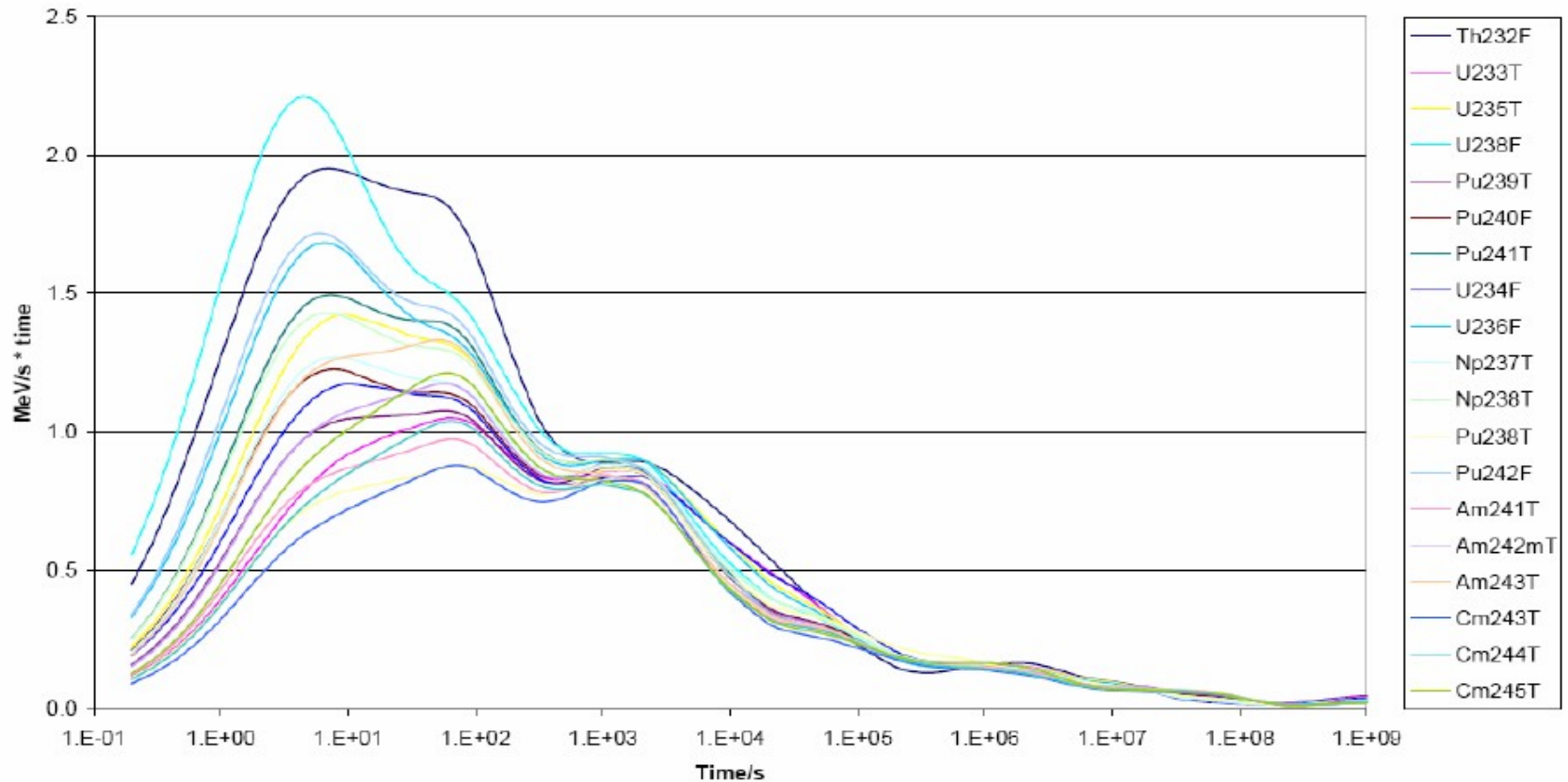
# Thermal UOX decay heat split FP+HE

- Decay heat from fission products and heavy elements ( $Z > 80$ )

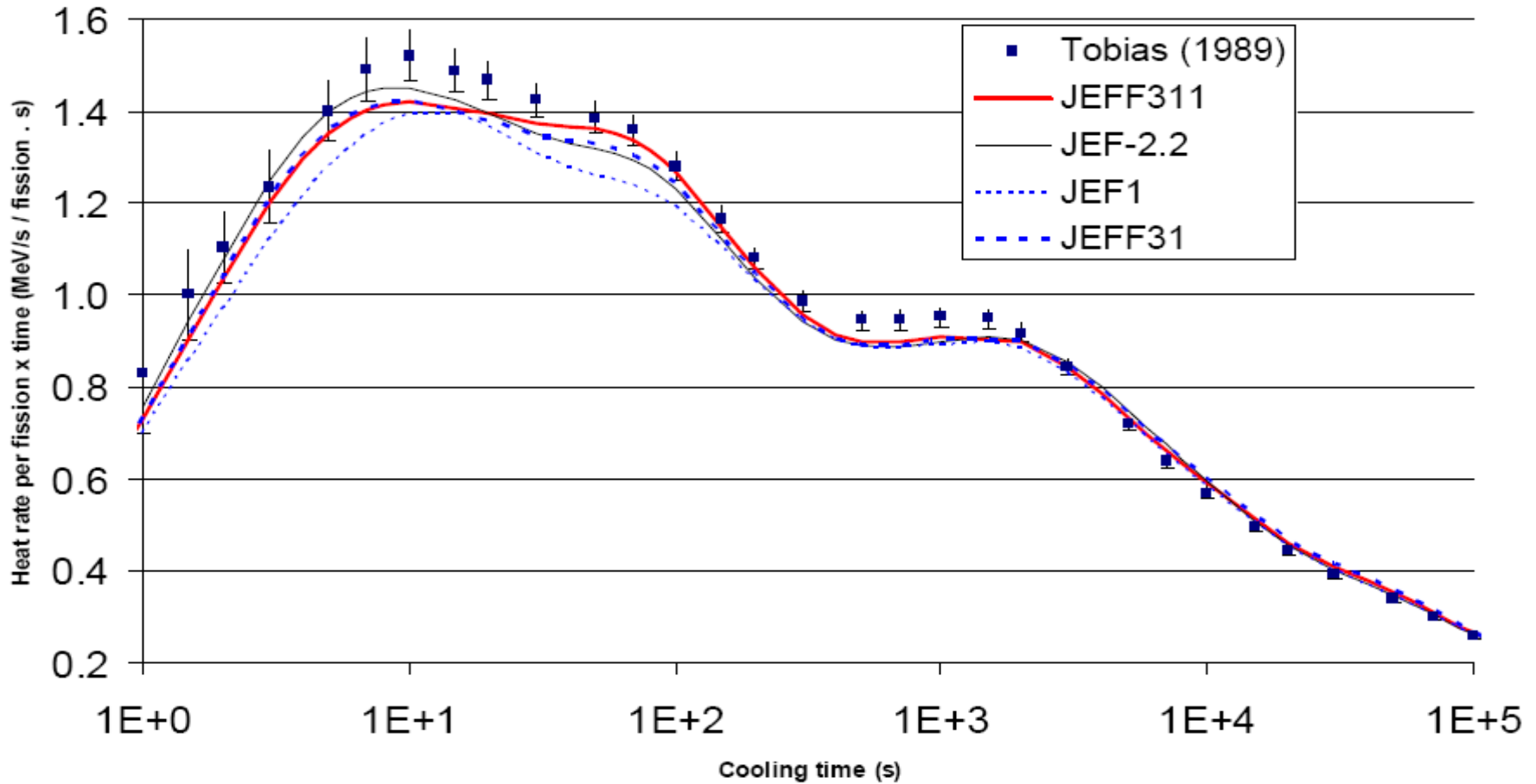


# Short term FP decay heat calculation by nuclide

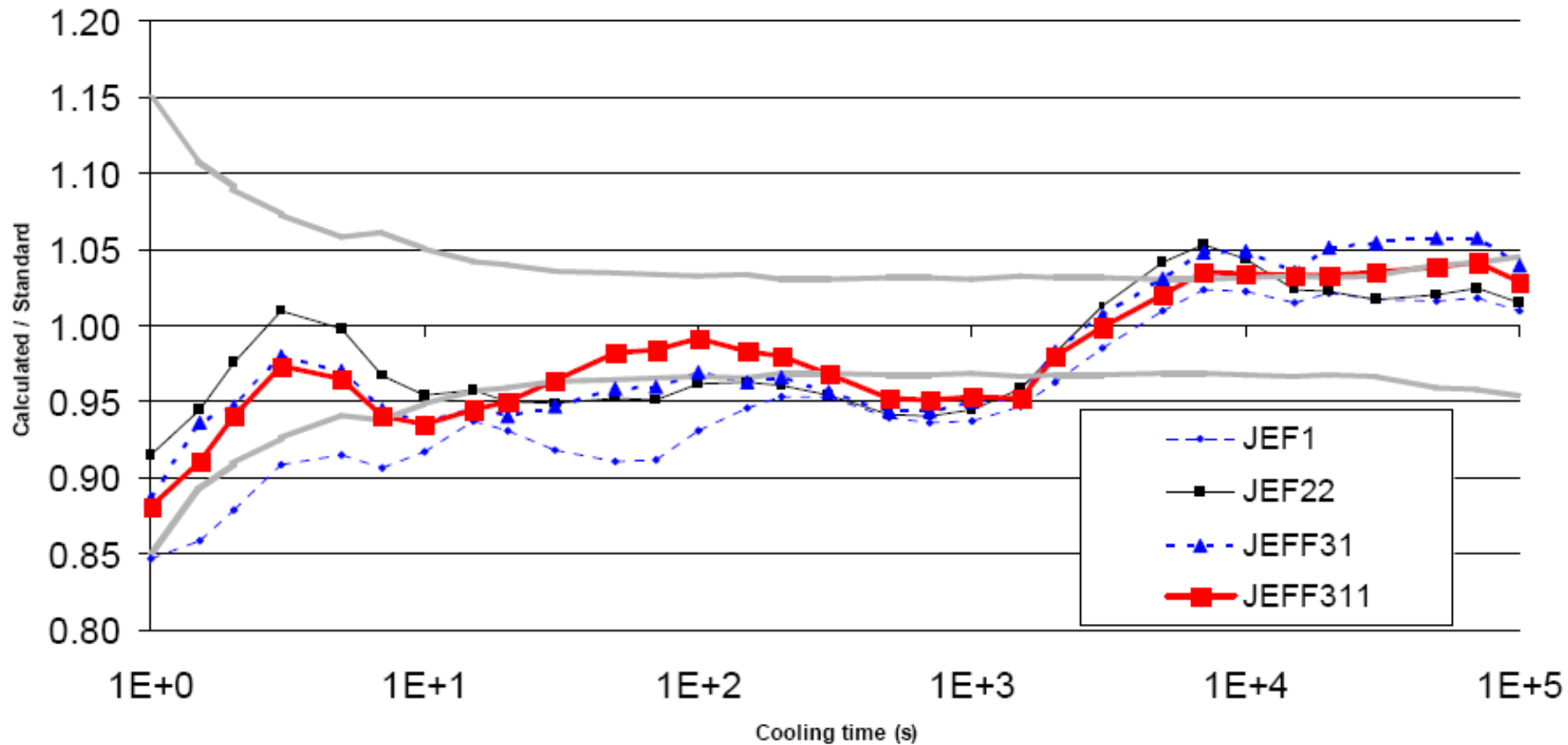
- decay heat from a single fission (JEFF\_3.1)



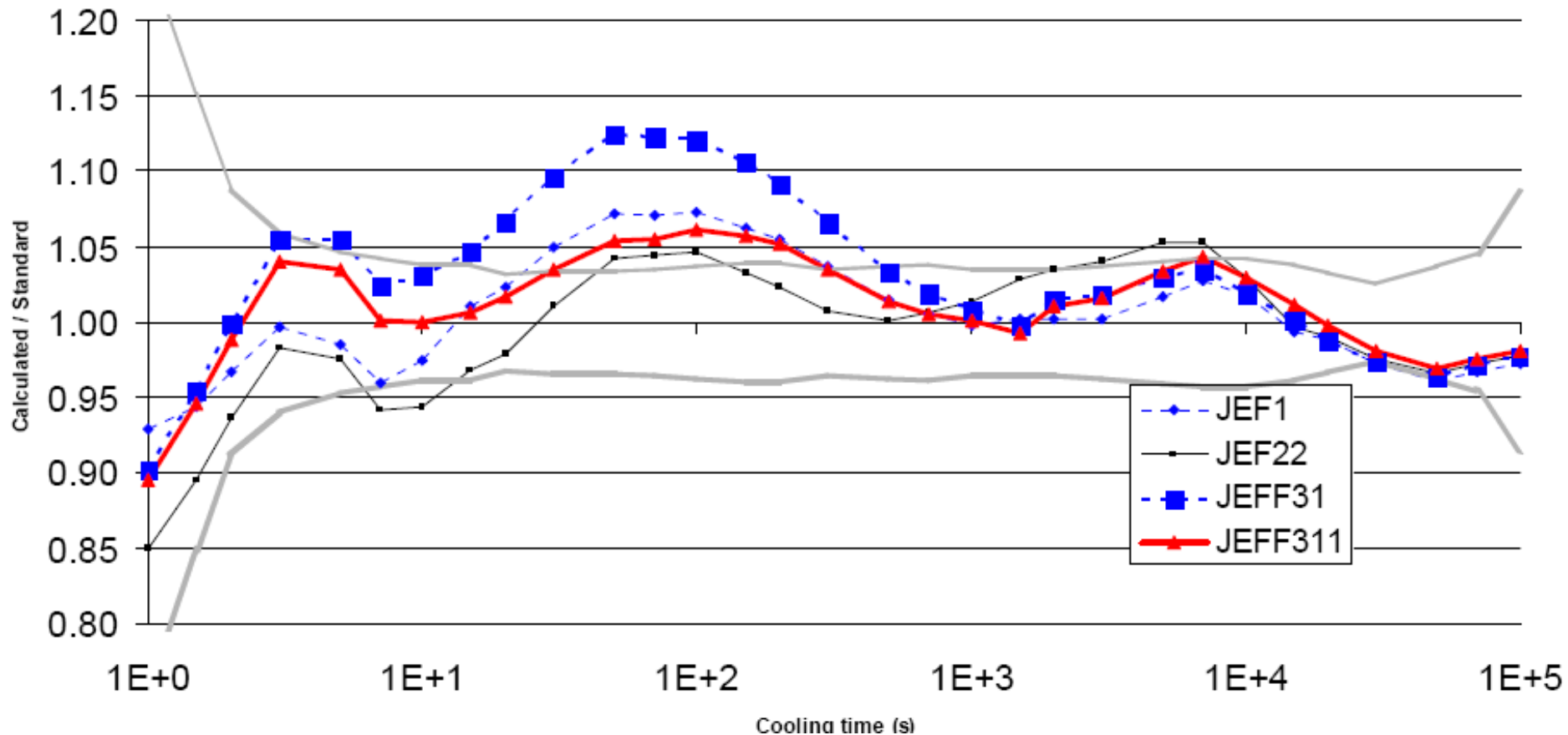
# Short term validation- U235 ( $\beta+\gamma$ )



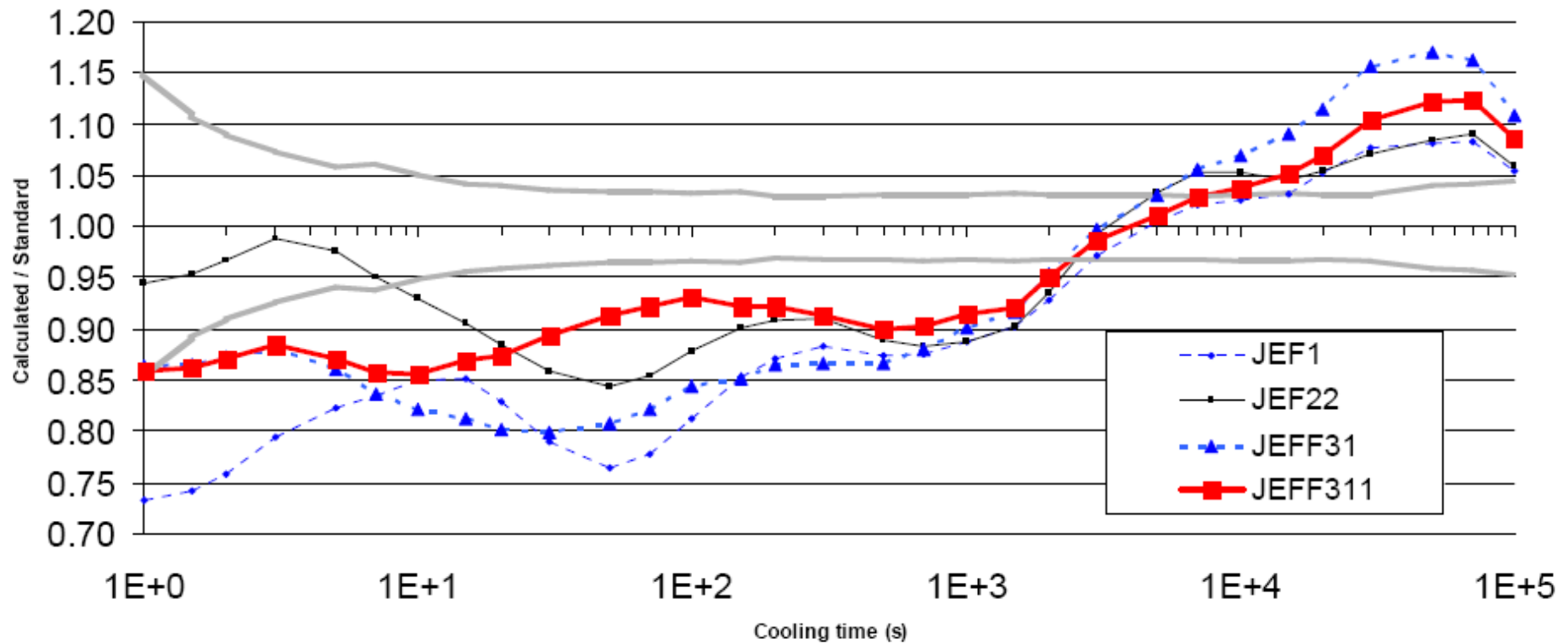
# Short term validation- U235 ( $\beta+\gamma$ )



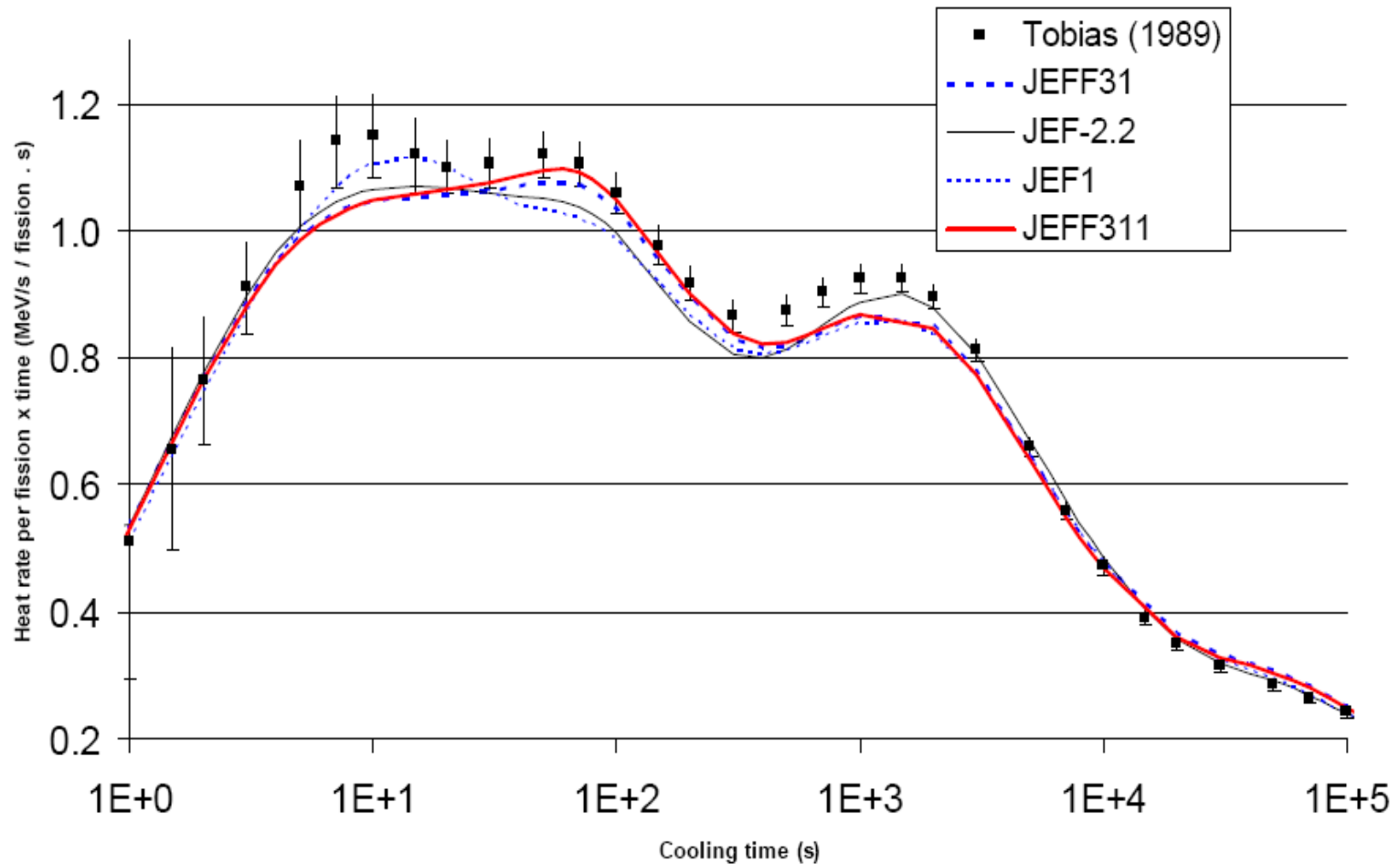
# Short term validation- U235 ( $\beta$ )



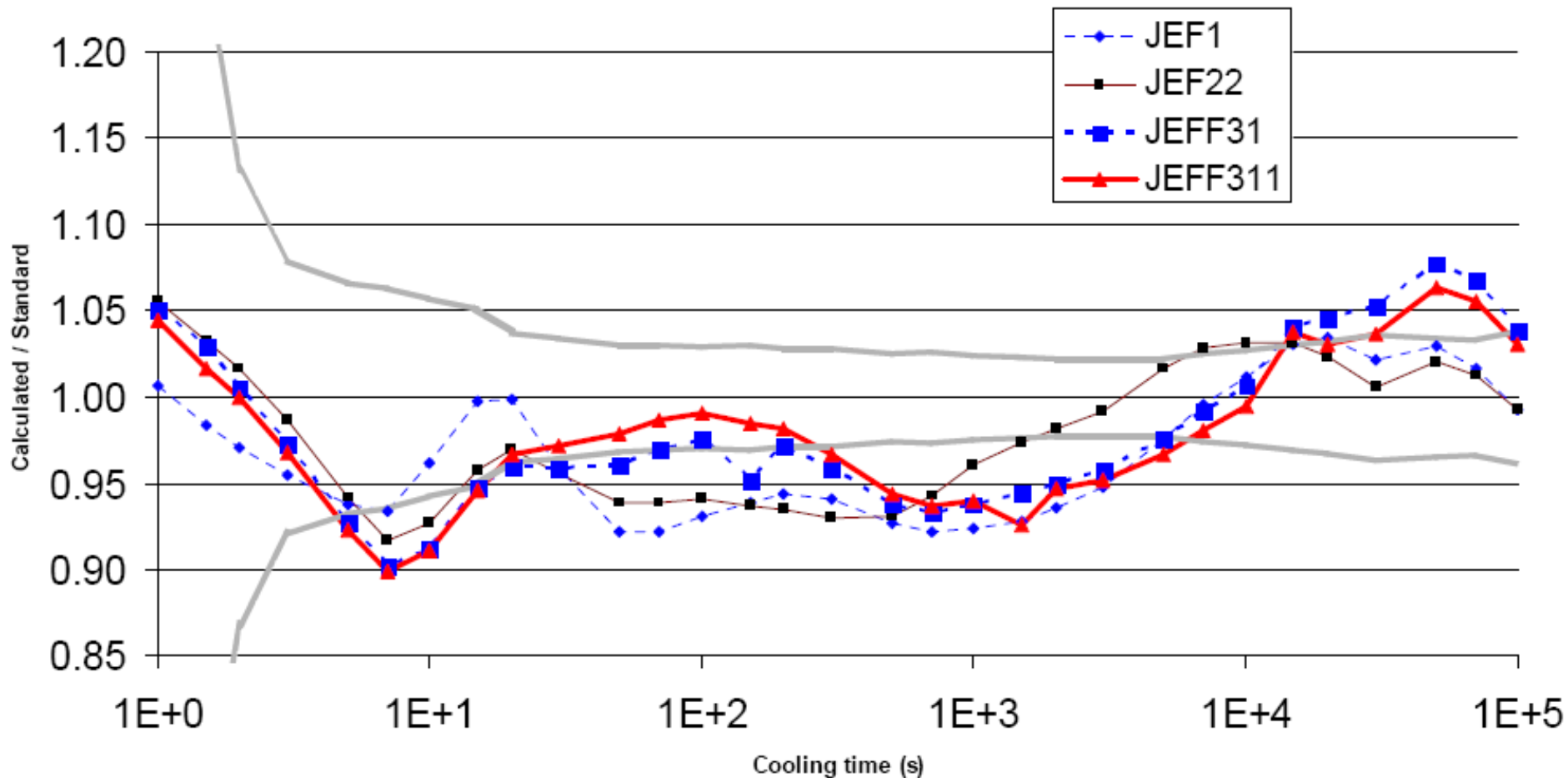
# Short term validation- U235 ( $\gamma$ )



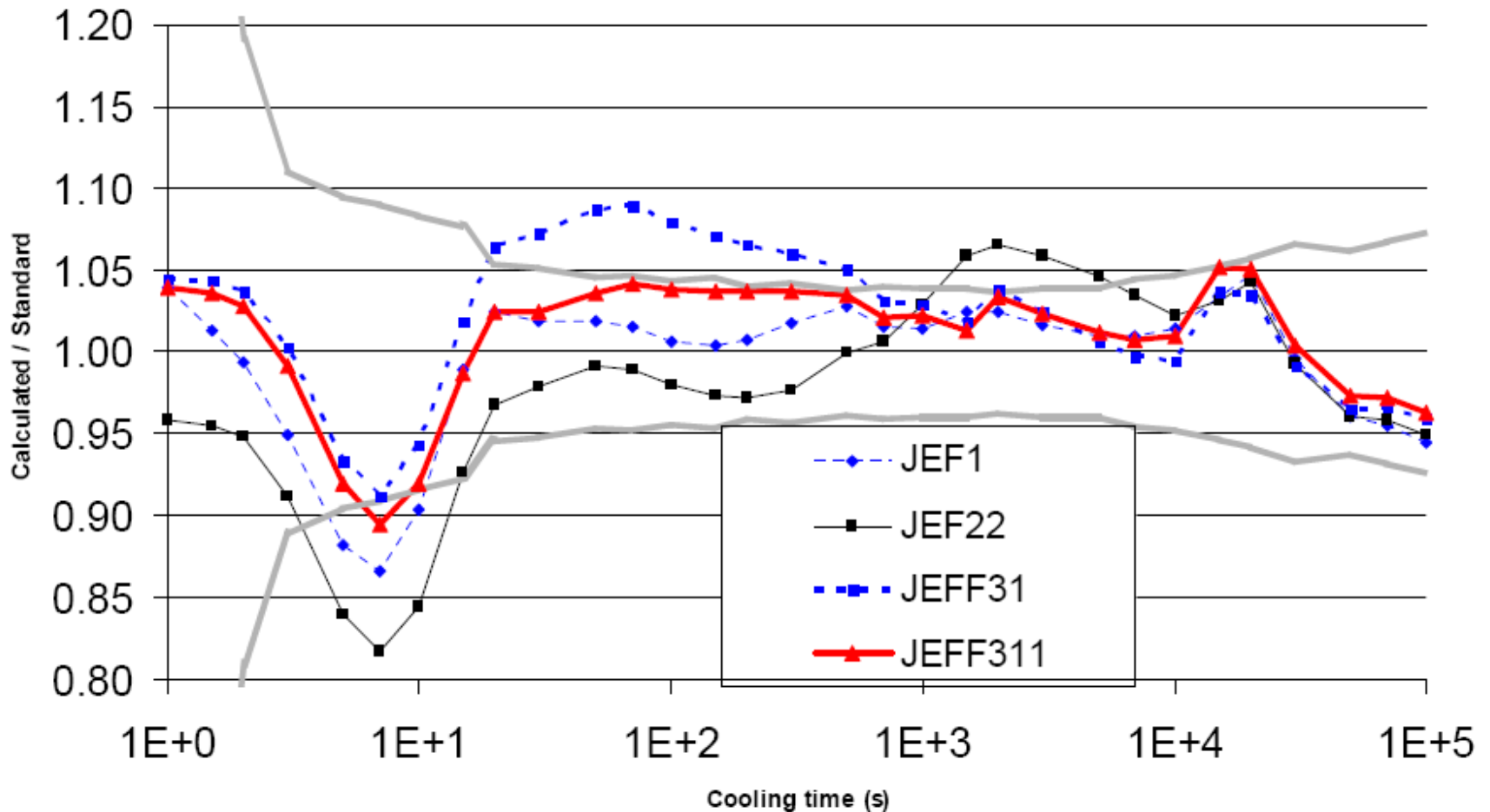
# Short term validation- Pu239 ( $\beta+\gamma$ )



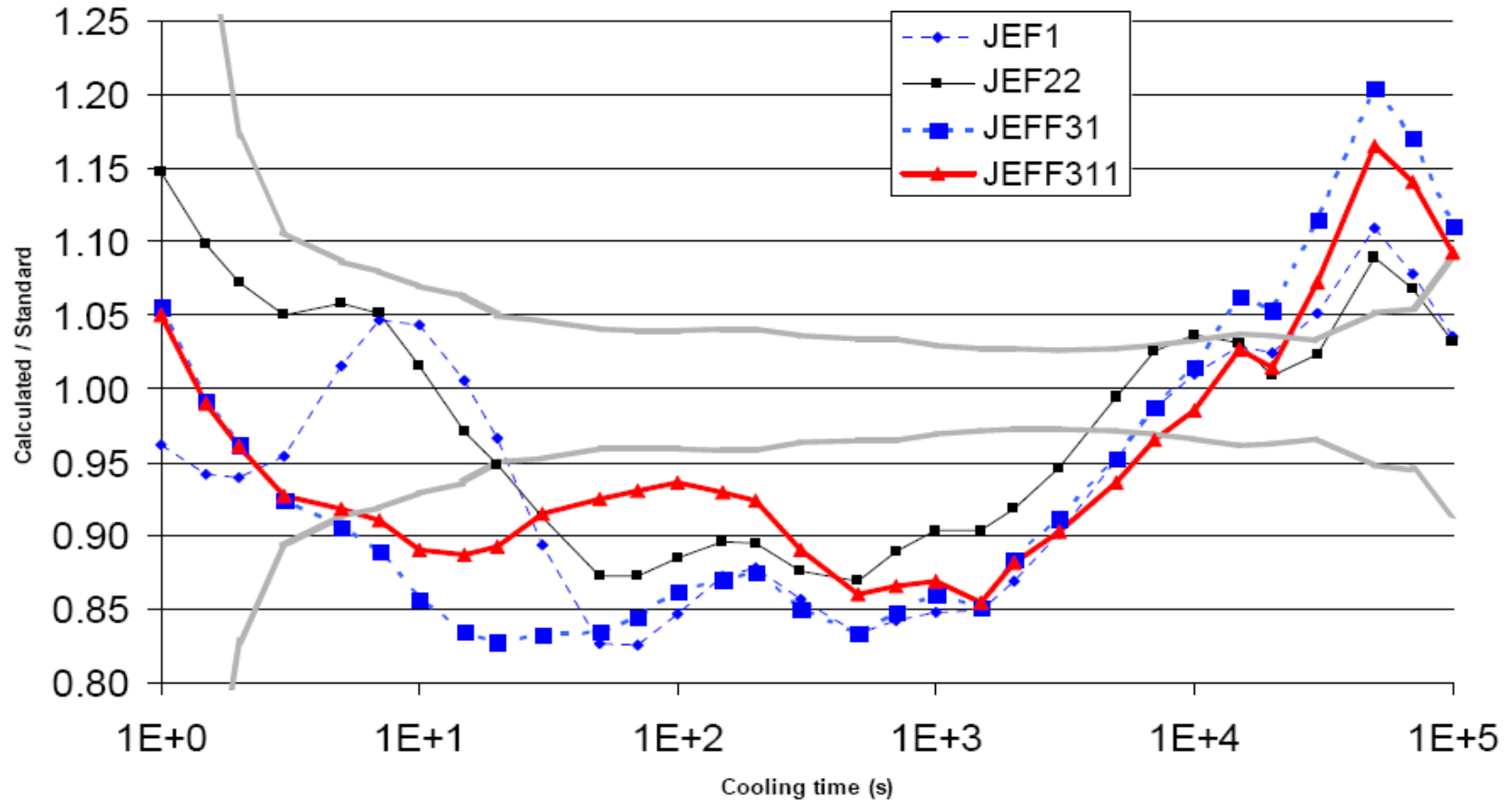
# Short term validation- Pu239 ( $\beta+\gamma$ )



# Short term validation- Pu239 ( $\beta$ )



# Short term validation- Pu239 ( $\gamma$ )



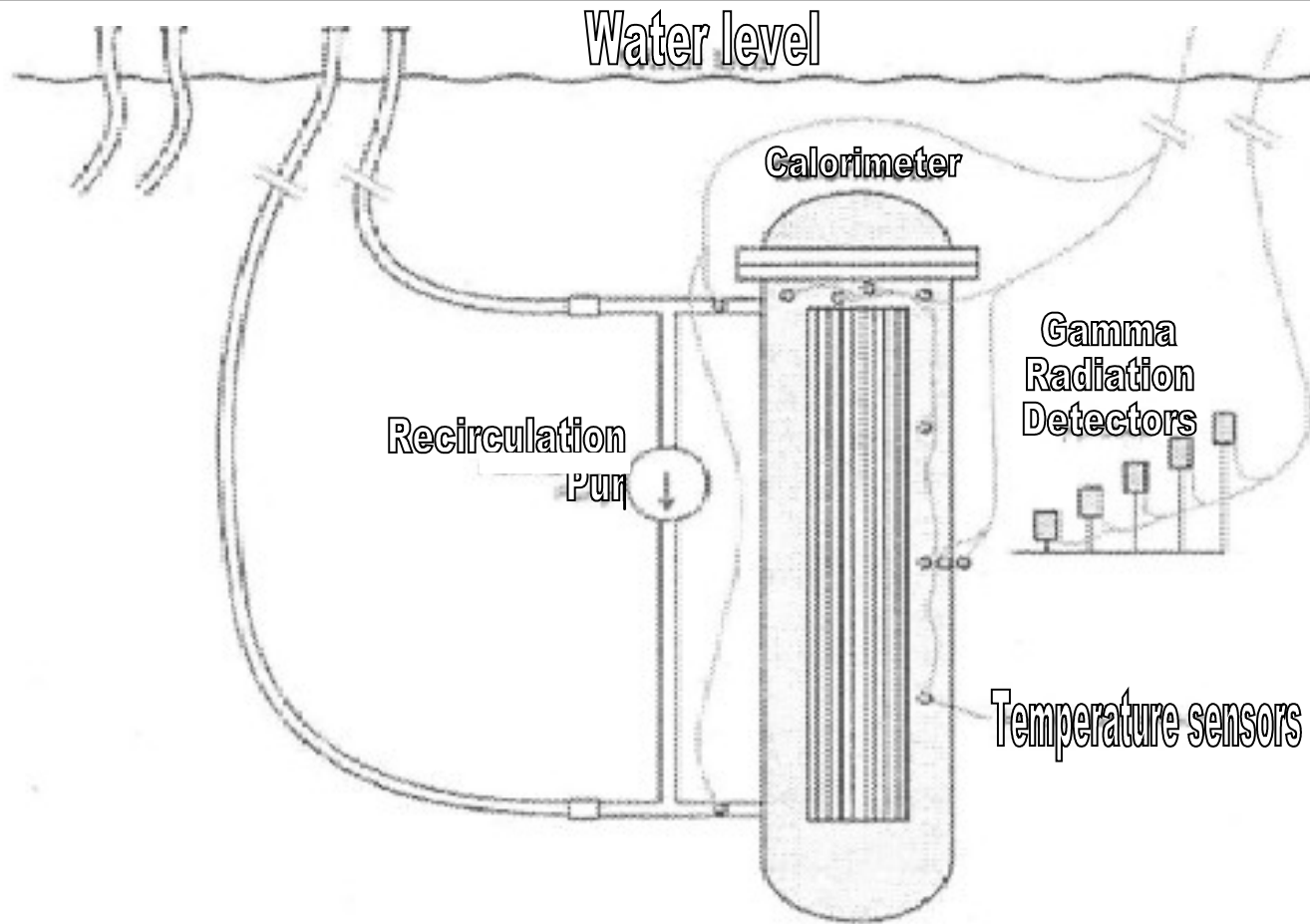
# Existing mid term decay heat validation

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Measurements of 24 fuel assemblies reported in F. Schmittroth, "ORIGEN2 Calculations of PWR spent fuel decay heat compare with calorimeter data." HEDL-TME 83-32 UC-85 (1984)

- Assemblies from 3 reactors;  
San Onofre, Point Beach and Turkey Point
- Whole assemblies measured.
- Burnup values of 25 to 40 GWd/t
- Cooling 2.4 to 8.2 years
- Enrichments 2.5 to 3.4% Wt% U235.
- Measurements are reported to be  $\pm 2\%$

# SKB Calorimeter design



# Problems with measurements

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- 4 results removed due to reported inconsistencies.
  - San Onofre stainless steel clad; no data on Co ppm
    - Expected to be between 120 and 1200 ppmThis gives mean C/E on decay heat of
    - 1.09 at 1200 ppm
    - 1.06 at 1000 ppm
    - 0.93 at 120 ppm
  - Thus unless cobalt content known can't justify use.
- Only 12 measurements could be used for validation.

# Existing decay heat validation

**Comparison of existing PWR assembly decay heat measurements with calculations using JEFF data.**

Reactor	Initial $^{235}\text{U}$ (Wt%)	Burnup (GWd/t)	Cooling (d)	Measured heat (W)	JEFF-2.2 C/E	JEFF-3.1 C/E
Point Beach	3.397	31.914	1635	724	0.98	0.97
		31.914	1635	723	0.98	0.97
		38.917	1634	921	1.01	1.00
		39.384	1633	931	1.01	1.00
		35.433	1630	846	0.97	0.96
		38.946	1629	934	1.00	0.99
		37.057	1630	874	1.00	0.99
Turkey Point	2.556	28.430	962	1423	1.05	1.04
		28.430	2077	625	1.03	1.01
		26.485	963	1284	1.06	1.05
		27.863	864	1550	1.06	1.05
	2.559	25.595	1782	637	1.00	0.98
Mean and Standard Deviation of C/E values				Point Beach	$0.99 \pm 0.02$	$0.98 \pm 0.02$
				Turkey Point	$1.04 \pm 0.03$	$1.02 \pm 0.03$
				Combined	$1.01 \pm 0.03$	$1.00 \pm 0.03$

# Recent published SKB decay heat measurements

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- Reference: Svensk Kärnbränslehantering AB, "Measurements of decay heat in spent nuclear fuel at the Swedish interim storage facility, Clab", SKB Report R-05-62, ISSN 1402-3091 (2006).
- Swedish Nuclear Fuel and Waste Management Company (SKB) have completed calorimetric measurements on BWR and PWR assemblies at the Swedish Interim Spent Fuel Storage Facility, CLAB, at Oskarshamn.
- 43 measurements of PWR assemblies and 66 measurements of BWR assemblies.
- Estimate of accuracy between 1 and 2%.

# Recent published SKB decay heat measurements

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- The PWR assemblies range in
  - irradiation from 19.7 to 51.0 GWd/t
  - cooling from 13 to 23 years and
  - enrichment from 2.1 to 3.4%.
  
- The BWR assemblies range in
  - irradiation from 14.5 to 46.6 GWd/t
  - cooling from 11 to 27 years and
  - enrichment from 2.1 to 3.1%.

# Recent published SKB decay heat measurements

- This considerably extends the current FISPIN decay heat validation.

- 12 measurements  $\Rightarrow$  121 measurements
- PWR  $\Rightarrow$  PWR and BWR
- Cooling 2.4 – 8.2 years  $\Rightarrow$  2.4 – 27 years
- Irradiation 25 – 40  $\Rightarrow$  14.5 – 51 GWd/t
- Enrichment 2.5 – 3.4 wt%  $\Rightarrow$  2.1 – 3.4 wt%

# Modelling assumptions

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- Published reactor design information
- 2D slice model through assembly
- WIMS/TRAIL/FISPIN route
- Neutron Cross sections from TRAIL database  
DB.WIMS172.6A\_S5
- Assumptions
  - PWR 500 ppm boron during whole irradiation
  - Structural materials irradiated in same flux level as fuel

# PWR results (1)

**Table 2: Comparison of decay heat from new SKB PWR assemblies with calculation.**

Assembly	Reactor	Enrichment (%U235)	Burnup (MWd/t)	Cooling (days)	Measured Decay Heat (W)	Calculated Decay Heat using JEF2.2 (W)	Calculated Decay Heat using JEF3.1.1 (W)	C/E values (JEF2.2)	C/E values (JEF3.1.1)
C01	Ringhals 2	3.10	36688	8468	415.8	435.63	430.00	1.048	1.034
C12		3.10	36385	8403	410.3	430.76	425.16	1.050	1.036
C20		3.10	35720	6950	415.8	447.93	442.35	1.077	1.064
				6951	426.1	447.88	442.35	1.051	1.038
				6952	428.9	447.88	442.35	1.044	1.031
				6959	435.6	447.70	442.17	1.028	1.015
C42		3.10	35639	5803	442.3	456.70	451.12	1.033	1.020
				5804	448.4	456.70	451.12	1.019	1.006
D27		3.25	39676	7669	456.1	472.00	465.91	1.035	1.022
D38		3.25	39403	8005	442.3	462.04	455.93	1.045	1.031
E38		3.20	33973	7999	376.3	388.45	382.87	1.032	1.017
E38				8000	374.3	388.45	382.87	1.038	1.023
E40		3.20	34339	8075	381.3	392.55	386.96	1.029	1.015
F14		3.20	34009	7722	381.3	397.15	391.40	1.042	1.026
F21		3.20	36273	7376	420.9	433.93	428.02	1.031	1.017
F25		3.20	35352	7725	396.7	416.33	410.48	1.049	1.035
F32		3.20	50962	5860	692	722.90	715.83	1.045	1.034
G11		3.19	35463	6990	416.4	428.81	422.89	1.030	1.016
G23		3.21	35633	6984	420.6	434.82	428.77	1.034	1.019
I09		3.20	40188	5849	507.9	534.98	527.92	1.053	1.039
I20	3.20	34313	6588	403.5	412.38	406.45	1.022	1.007	
I24	3.20	34294	6601	410.1	417.41	411.40	1.018	1.003	
I25	3.20	36859	6198	445.8	467.35	461.24	1.048	1.035	



# PWR results (2)

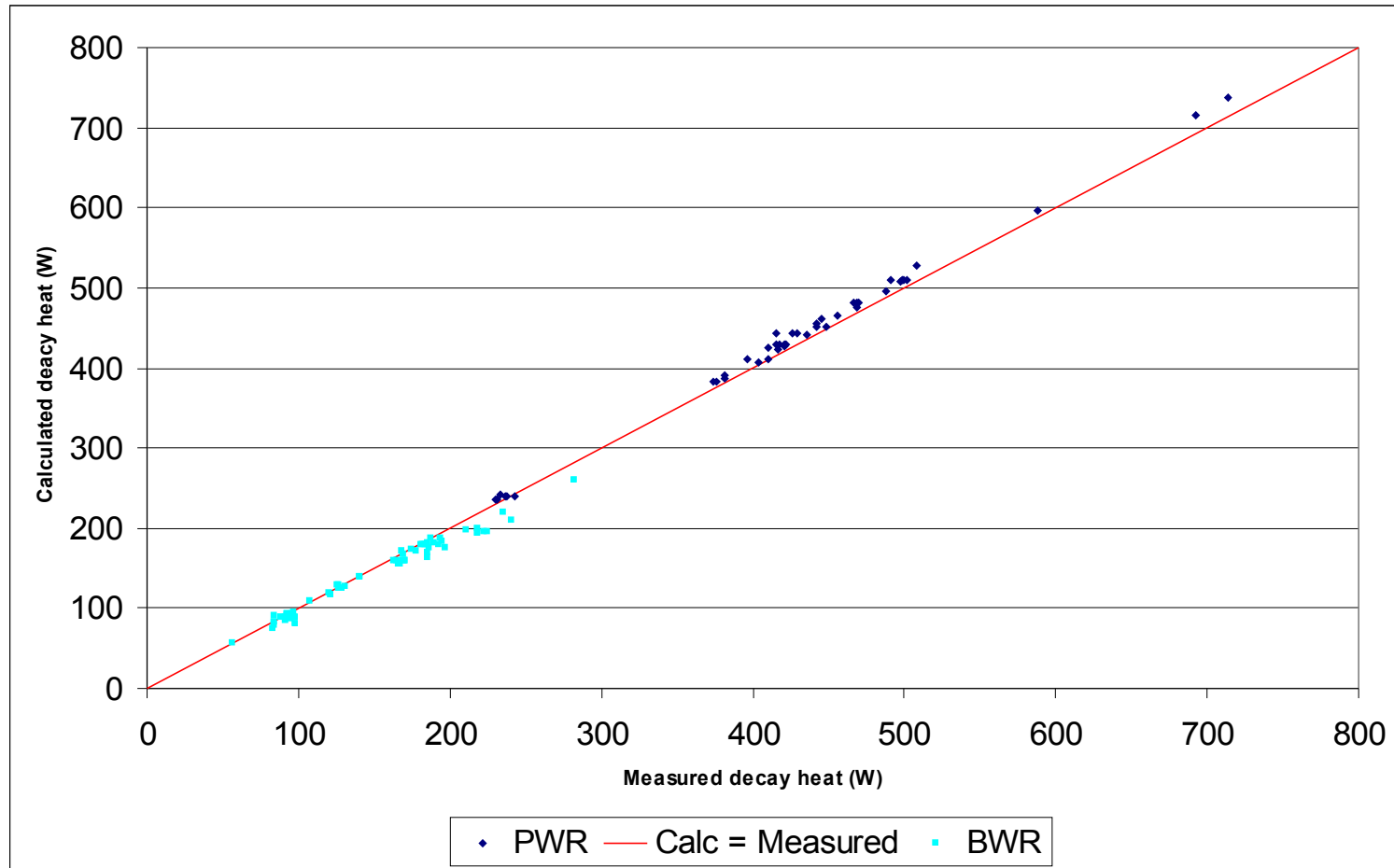
**Table 2: Comparison of decay heat from new SKB PWR assemblies with calculation.**

Assembly	Reactor	Enrichment (%U235)	Burnup (MWd/t)	Cooling (days)	Measured Decay Heat (W)	Calculated Decay Heat using JEF2.2 (W)	Calculated Decay Heat using JEF3.1.1 (W)	C/E values (JEF2.2)	C/E values (JEF3.1.1)
0C9	Ringhals 3	3.10	38442	6551	491.2	516.29	509.83	1.051	1.038
0E2		3.10	41628	5823	587.9	603.26	596.23	1.026	1.014
0E6		3.10	35993	5829	487.8	502.97	496.44	1.031	1.018
1C2		3.10	33318	6559	417.7	436.12	429.97	1.044	1.029
1C5		3.10	38484	6593	499.2	516.23	509.77	1.034	1.021
1E5		3.10	34638	5818	468.8	483.01	475.99	1.030	1.015
2A5		2.10	20107	7297	233.8	245.23	241.55	1.049	1.033
2C2		3.10	36577	6550	466.5	488.51	482.03	1.047	1.033
3C1		3.10	36572	6545	470.2	487.39	480.92	1.037	1.023
3C4		3.10	38447	6544	497.3	515.09	508.65	1.036	1.023
3C5		3.10	38373	6543	501.4	516.77	510.29	1.031	1.018
3C9		3.10	36560	6552	468.4	487.68	481.20	1.041	1.027
4C4		3.10	33333	6572	422	436.16	430.01	1.034	1.019
4C7		3.10	38370	6550	498.7	516.07	509.60	1.035	1.022
5A3		2.10	19699	6972	237.7	243.23	239.38	1.023	1.007
5A3		2.10		6975	236.6	243.19	239.38	1.028	1.012
5A3		2.10		6977	243.4	243.19	239.33	0.999	0.983
5A3		2.10		7291	230.9	239.72	235.96	1.038	1.022
5A3		2.10		7304	230.2	239.58	235.82	1.041	1.024
5F2	3.40	47308		4724	714.1	736.05	737.97	1.031	1.033

# PWR comparison

Reactor / #	JEF-2.2 (C/E)	JEFF-3.1.1 (C/E)
Point Beach / 7	$0.99 \pm 0.02$	$0.98 \pm 0.02$
Turkey Point / 5	$1.04 \pm 0.03$	$1.02 \pm 0.03$
Ringhals 2 / 23	$1.039 \pm 0.013$	$1.025 \pm 0.014$
Ringhals 3 / 20	$1.034 \pm 0.011$	$1.021 \pm 0.012$

# Decay Heat Calculated against Experimental



# Benchmark Document available from NNL

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- Report describes
  - reactor design
  - assembly design
  - irradiation information
  - assumptions required to complete models
- Should allow others to repeat validation and
  - review input data used
  - review methods used
  - report differences in results from other methods.

# Methods of estimating decay heat accuracy

Can estimate decay heat biases and uncertainties by

- Comparing measurements of heating to calculation, or
- Error propagation from radionuclide biases and uncertainties

Expected heat from nuclide  $i$ ,  $h_i = N_i(\text{expected}) \lambda_i E_i$

where  $N_i(\text{expected}) = N_i(\text{FISPIN}) / (\text{C/E bias of FISPIN for } i)$

Expected Heat,  $H = \sum h_i$  (if considering chemical separation can still be applied, with chemical df's)

Assumptions:

- If no PIE results, assume no C/E bias (i.e = 1) but 100% error.
- If nuclide is a daughter of one measured assumed same bias and C/E.

# Post irradiation validation of Gösgen samples (ARIANE)

Nuclide	Sample C/E				Mean ; SD
	GU1	GU3'	GU3	GU4	
Sr90	0.77	1.03	0.98	0.99	0.94 ; 0.12
Mo95	1.00	0.88	0.94	0.97	0.95 ; 0.05
Tc99	1.03	0.91	1.03	1.27	1.06 ; 0.15
Ru101	1.06	0.87	0.95	0.97	0.96 ; 0.08
Ru106	1.08	0.87	0.47	0.85	0.82 ; 0.25
Rh103	1.14	1.16	1.19	0.96	1.11 ; 0.10
Ag109	2.18	1.09			1.64 ; 0.77
Sb125	1.90	1.98			1.94 ; 0.05
I129		0.99	0.97	0.90	0.96 ; 0.05
Cs133	1.02	1.04	0.95	0.99	1.00 ; 0.04
Cs134	1.05	1.03	0.84	1.01	0.98 ; 0.10
Cs135	1.05	1.07	1.01	1.14	1.07 ; 0.05
Cs137	0.97	0.99	0.95	1.06	0.99 ; 0.05
Ce144	1.06	1.07	1.06	1.08	1.07 ; 0.01
Nd142	0.93	0.97	1.02	1.11	1.01 ; 0.08
Nd143	1.14	1.05	1.11	0.98	1.07 ; 0.07
Nd144	0.95	0.97	0.97	0.94	0.95 ; 0.02
Nd145	1.01	1.00	1.03	0.98	1.00 ; 0.02
Nd146	1.00	0.99	0.99	0.98	0.990 ; 0.008
Nd148	1.00	1.00	1.00	1.00	1.000 ; 0.000
Nd150	0.98	0.98	0.93	1.01	0.97 ; 0.03
Pm147	1.46	1.16	0.85	0.94	1.10 ; 0.27
Sm147	0.94	1.02	0.98	1.05	1.00 ; 0.05
Sm148	0.91	0.90	0.82	1.00	0.91 ; 0.07
Sm149	1.16	1.37	0.97	1.14	1.16 ; 0.16
Sm150	1.03	1.06	1.01	1.07	1.04 ; 0.03
Sm151	1.34	1.30	1.25	1.26	1.29 ; 0.04

Nuclide	Sample C/E				
	GU1	GU3'	GU3	GU4	
Sm152	1.02	1.15	1.14	1.12	1.11 ; 0.06
Sm154	0.97	1.08	1.09	1.12	1.07 ; 0.07
Eu151	0.57	0.76			0.67 ; 0.14
Eu153	1.13	1.11	1.03	1.10	1.09 ; 0.04
Eu154	2.27	1.80	1.57	1.60	1.81 ; 0.32
Eu155	1.05	0.99	0.86	0.96	0.96 ; 0.08
Gd155	1.12	1.23	1.00	0.68	1.01 ; 0.24
U234	1.17	1.39	1.43	1.39	1.35 ; 0.12
U235	1.41	1.14	1.26	1.06	1.21 ; 0.15
U236	1.02	1.01	0.99	1.01	1.01 ; 0.01
U238	1.00	1.00	1.00	0.99	0.996 ; 0.001
Np237		0.90	0.84	0.74	0.82 ; 0.08
Pu238	1.03	0.97	0.90	1.04	0.98 ; 0.06
Pu239	1.18	1.04	1.04	1.07	1.08 ; 0.07
Pu240	1.02	0.98	0.96	0.99	0.99 ; 0.03
Pu241	1.15	1.09	1.06	1.09	1.10 ; 0.03
Pu242	0.92	1.00	0.91	1.02	0.96 ; 0.06
Pu244	0.69	0.55			0.62 ; 0.10
Pu	1.09	1.03	1.01	1.05	1.05 ; 0.04
Am241	1.23	1.28	1.25	1.06	1.20 ; 0.10
Am242m	1.24	0.93			1.08 ; 0.22
Am243	1.01	1.08	0.83	1.08	1.00 ; 0.12
Cm242	0.98	0.93			0.96 ; 0.03
Cm243	3.13	1.18			2.16 ; 1.38
Cm244	0.89	0.84	0.57	0.81	0.78 ; 0.14
Cm245	1.05	0.87	0.57	0.87	0.84 ; 0.20
Cm246	0.68	0.73			0.71 ; 0.03



# Decay heat for PWR PIE results

Calculation for PWR UOX 5%  $^{235}\text{U}$  enriched, irradiated to 40 GWd/t.

Cooling time (Years unless stated)	FISPIN heat (kW/tUi)	Expected bias (%)	Expected uncertainty (%)	Fraction of heat from measured nuclides (%)	Expected heat (kW/tUi)	Expected heat uncertainty (kW/tUi)
Shutdown	2.45E+03	0.26	93.49	7.83	2.45E+03	2.29E+03
3 days	1.58E+02	0.74	49.43	52.43	1.56E+02	7.73E+01
5	2.18E+00	-0.31	4.76	99.35	2.19E+00	1.04E-01
10	1.38E+00	0.50	4.79	99.31	1.37E+00	6.58E-02
15	1.19E+00	0.27	4.74	99.42	1.18E+00	5.61E-02
20	1.07E+00	0.23	4.58	99.54	1.07E+00	4.89E-02
25	9.74E-01	0.35	4.43	99.63	9.71E-01	4.30E-02
30	8.93E-01	0.62	4.31	99.71	8.87E-01	3.82E-02
100	3.50E-01	7.32	4.79	100.00	3.27E-01	1.56E-02
1000	6.48E-02	12.35	5.82	100.00	5.77E-02	3.36E-03
50000	3.39E-03	7.05	6.31	99.56	3.17E-03	2.00E-04
100000	1.26E-03	5.07	5.06	98.78	1.20E-03	6.08E-05
1000000	4.58E-04	-8.59	4.90	97.87	5.02E-04	2.46E-05

# Results

- Technique gives large uncertainties where decay heat is not dominated by measured nuclides.
- Uncertainty  $\sim 5\%$  from this general technique is larger than PWR assembly heat validation  $\sim 1-2\%$ .
- Bias less than  $1\%$  when fission products dominate,  $\leq 80$  years.
- When actinides dominate, the large biases for  $^{241}\text{Am}$  ( $20\%$ ) and  $^{239}\text{Pu}$  ( $+8\%$ ) lead to a over-prediction in heat of up to  $12\%$  at 1000 years.
- At  $1 \times 10^6$  years, the bias on  $^{237}\text{Np}$  ( $-18\%$ ) leads to a  $9\%$  under-prediction of heat.
- **This analysis can only be carried out if PIE exists, for fast and ADS systems such data is not available.**

# UOX and MOX case comparisons

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Thermal MOX decay heat is known to be greater than thermal UOX due to higher minor actinide concentrations but

- By how much?
- What uncertainty?

ARIANE included MOX samples

Thus can apply a similar approach to a MOX case

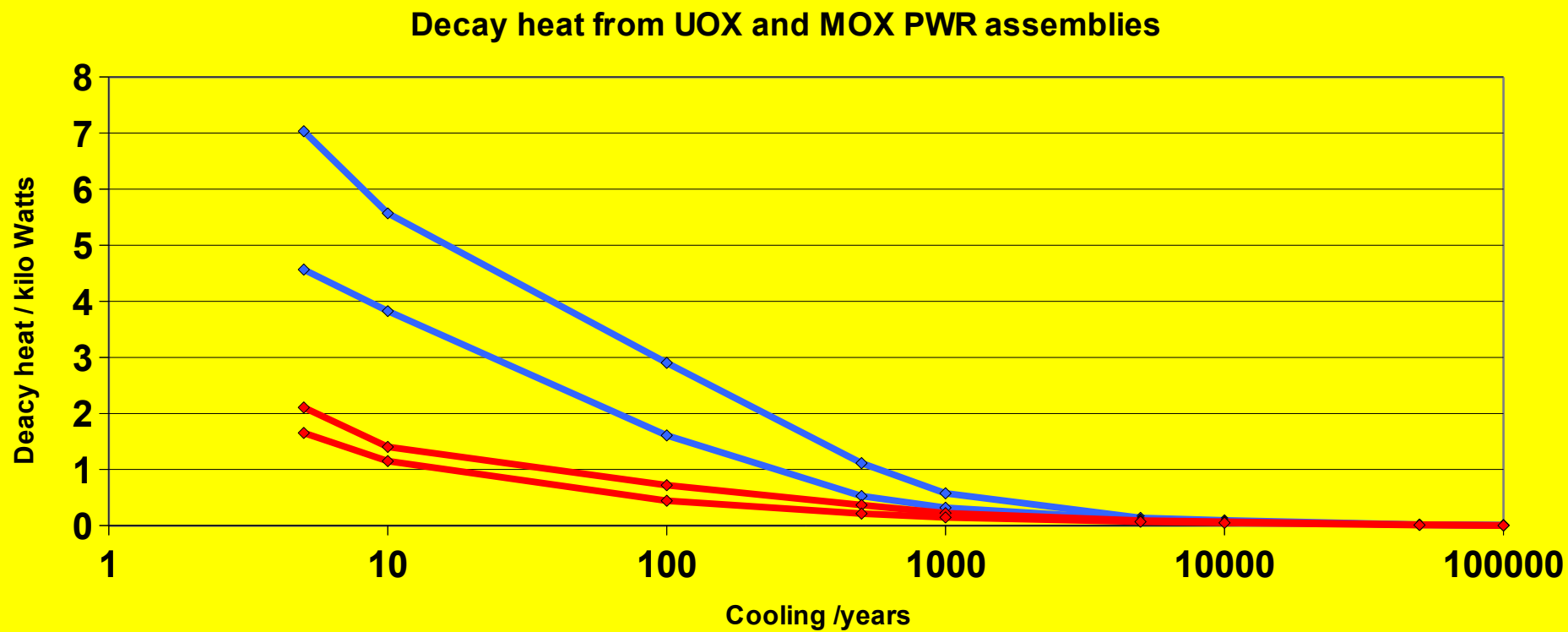
Considering equivalent UOX and MOX cases

To 50GWd/t in 5 cycles at 35 MW/t.

# MOX cases

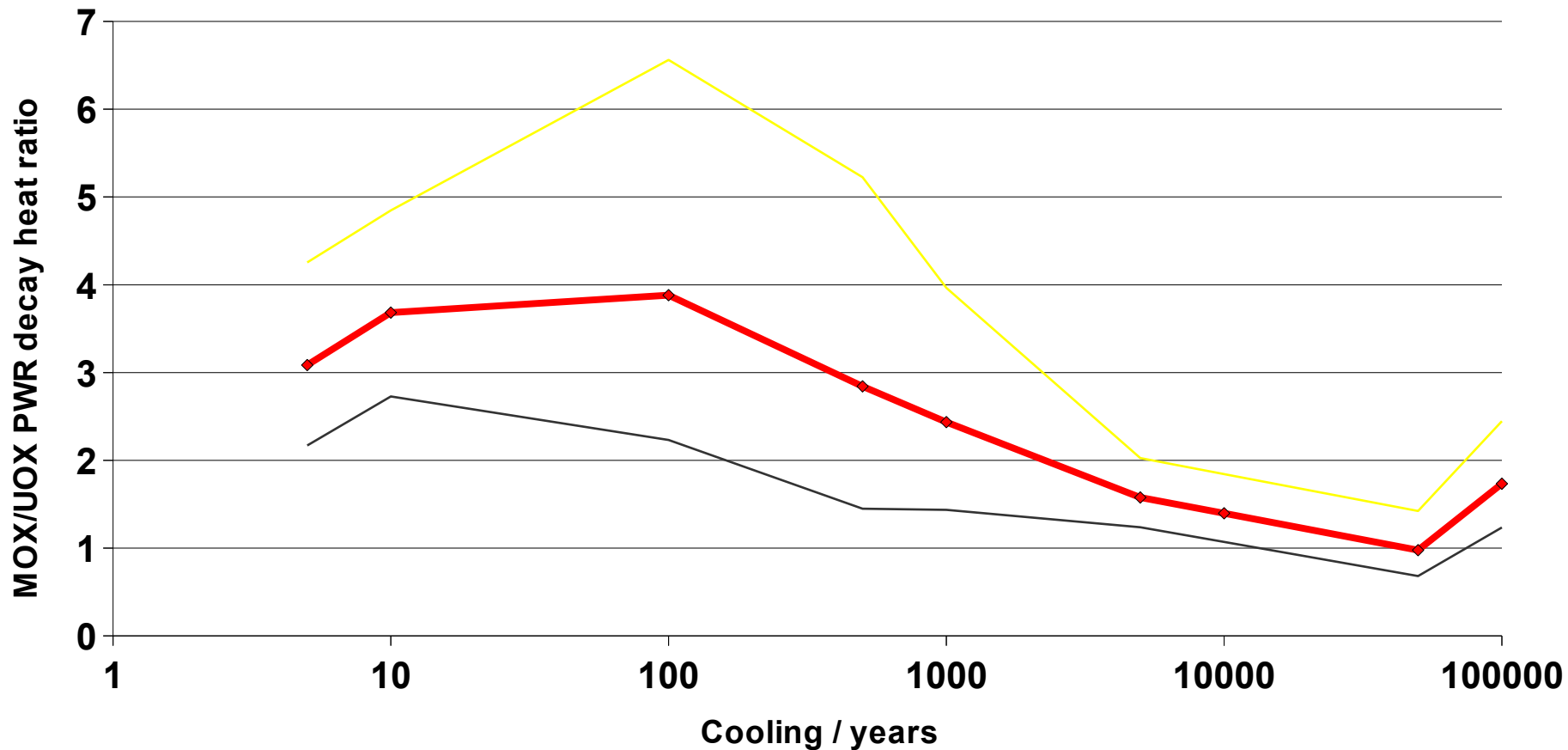
*Preliminary*

Years cooling	UOX heat /W	% Bias	% err 1 sd	MOX heat /W	% Bias	% err 1 sd	MOXheat / UOX heat
0	2071.513	-0.6	88.7	2034.080	94.1	178.0	0.98
5	1.845	1.8	5.1	5.218	5.1	12.0	3.09
10	1.292	-1.3	4.3	4.257	4.1	9.9	3.68
100	0.651	-10.8	6.5	2.150	11.9	23.8	3.88
500	0.331	-12.4	6.9	0.805	16.7	26.2	2.84
1000	0.204	-10.1	5.4	0.441	13.8	20.9	2.44
5000	0.082	-4.4	4.6	0.126	4.3	13.7	1.58
10000	0.063	-5.1	5.2	0.086	4.5	15.4	1.40
50000	0.015	-6.9	6.8	0.014	6.6	20.4	0.98
100000	0.004	-5.1	6.3	0.006	4.5	17.7	1.73



# MOX/UOX heat ratio

*Preliminary*



# Differences in decay heat contributions *Preliminary*

## UOX

- 5 years

Ba137m	22.8% (7.1E4)
Y90	18.5%
Ru106	13.3%
Cs134	8.0%

- 10 years

Ba137m	28.9% (6.3E4)
Y90	23.4%
Am241	15.1%

## MOX

- 5 years

Pu238	29.1%
Cm244	27.2%
Ba137m	7.9% (7.0E4)

- 10 years

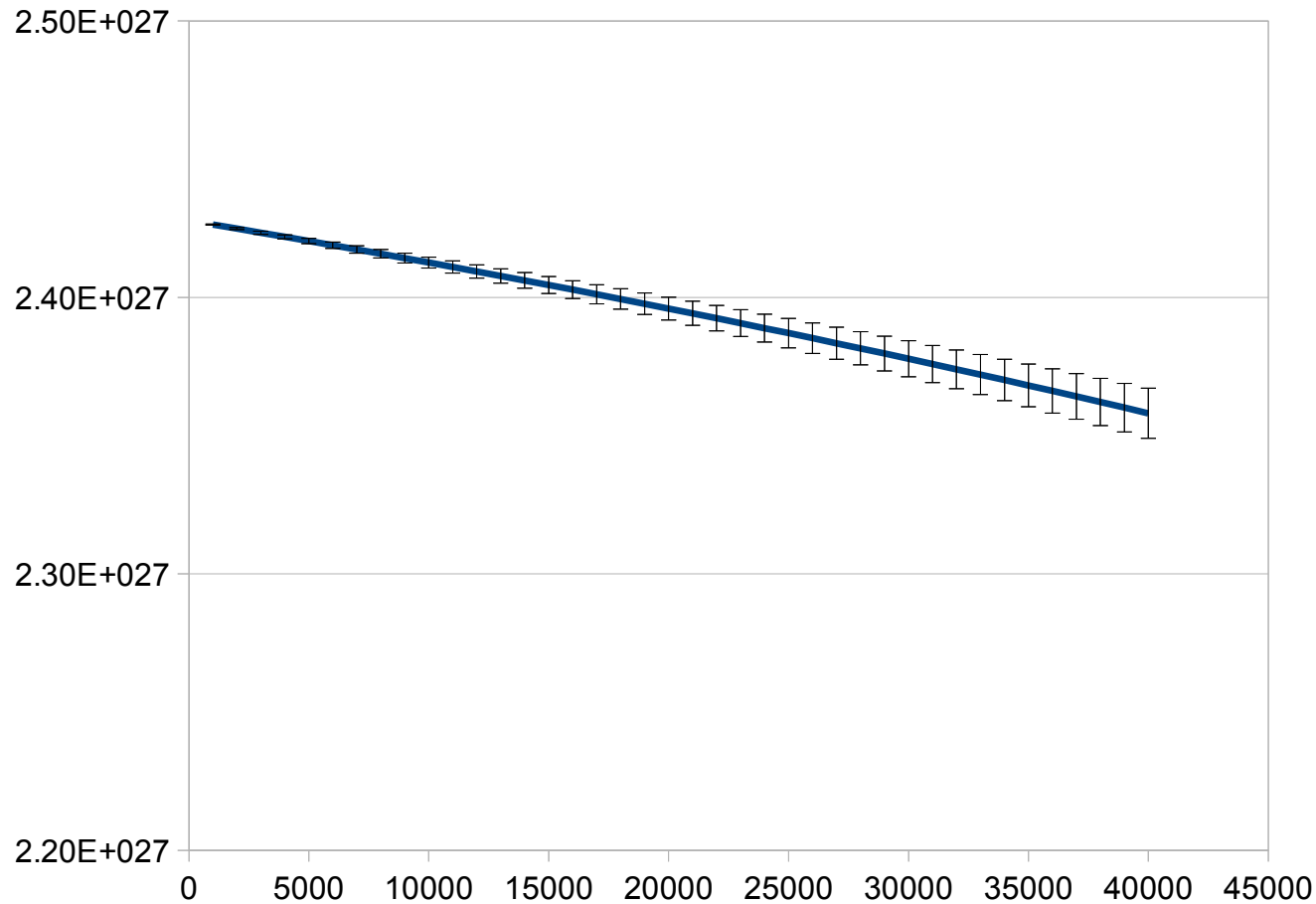
Pu238	34.3%
Cm244	27.5%
Am241	12.7%
Ba137M	8.7% (6.2E4)



# More rigorous method?

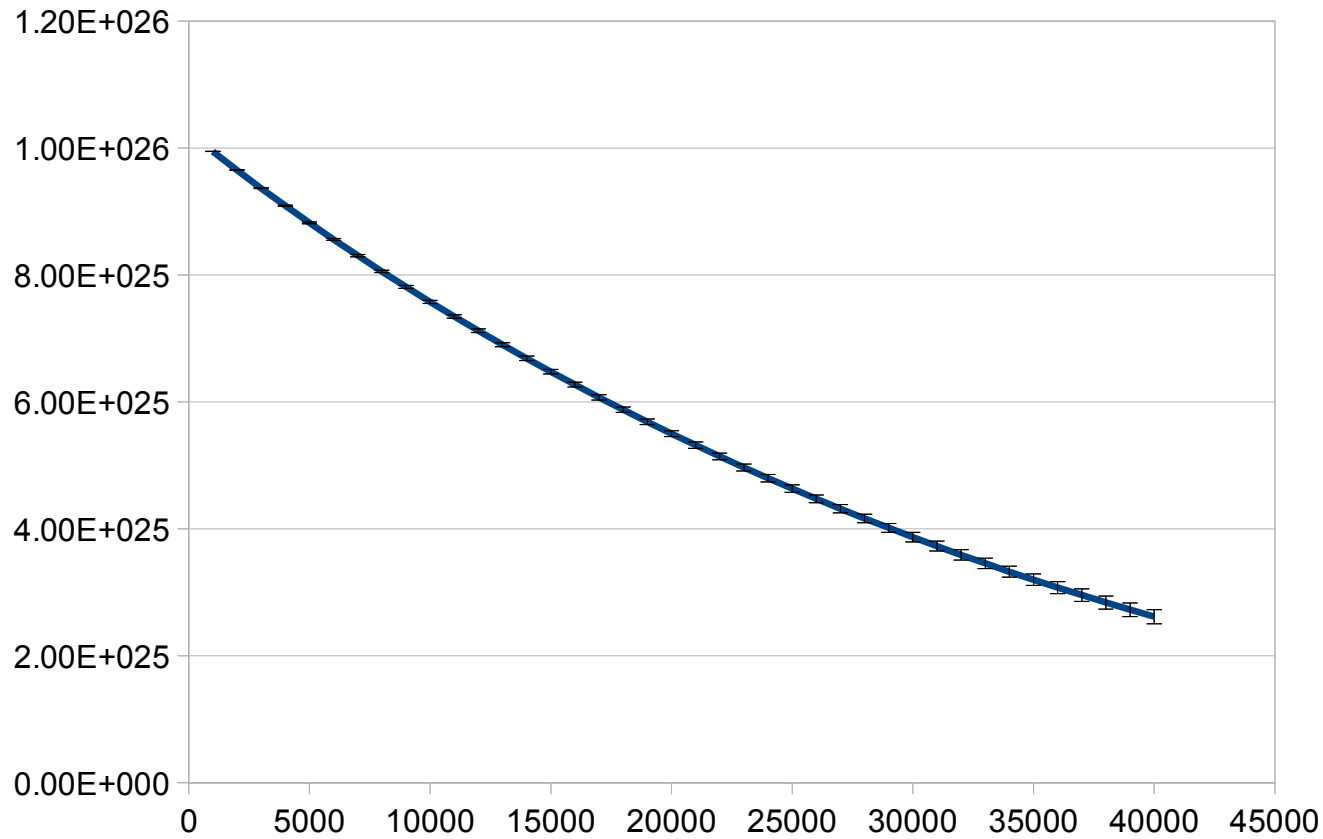
- The above is a semi-empirical method based upon post-irradiation measurements
  - Only valid if sufficient PIE is available
- Can we estimate the uncertainties by error propagation
- In the following slides a simple calculation is carried out using the following assumptions
  - Only capture and fission cross-sections important
  - Fluxes and cross-sections taken from FISPIN
  - Calculated number densities to within  $\pm 1\%$  of FISPIN
  - Uncertainties on cross-sections based upon Mughabghab (2006) or “typical” measurements

# Simple UOX calculation-U238 *Preliminary*



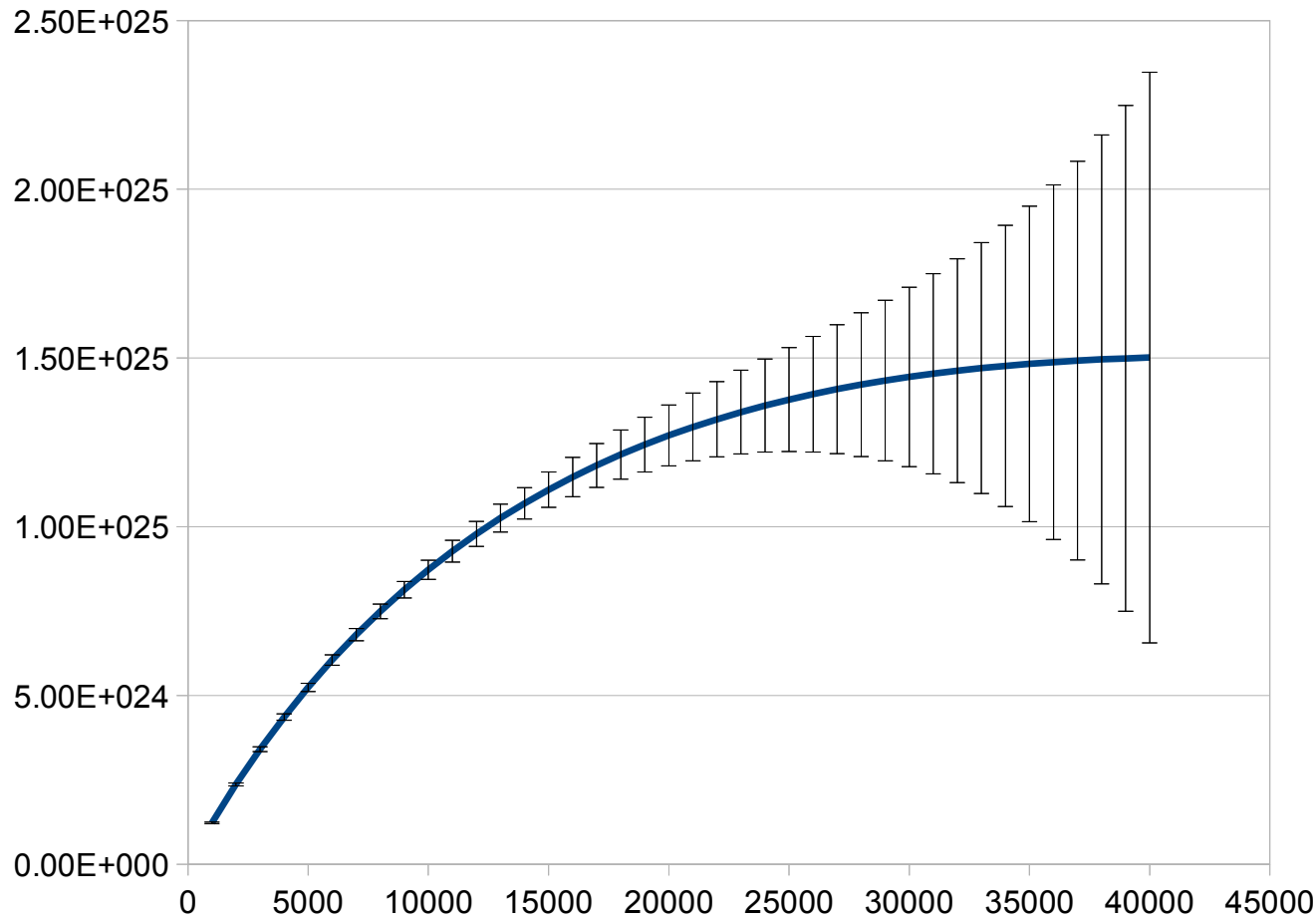
At end  
± 0.05 %

# Simple UOX calculation-U235 *Preliminary*



At end  
 $\pm 4.3 \%$

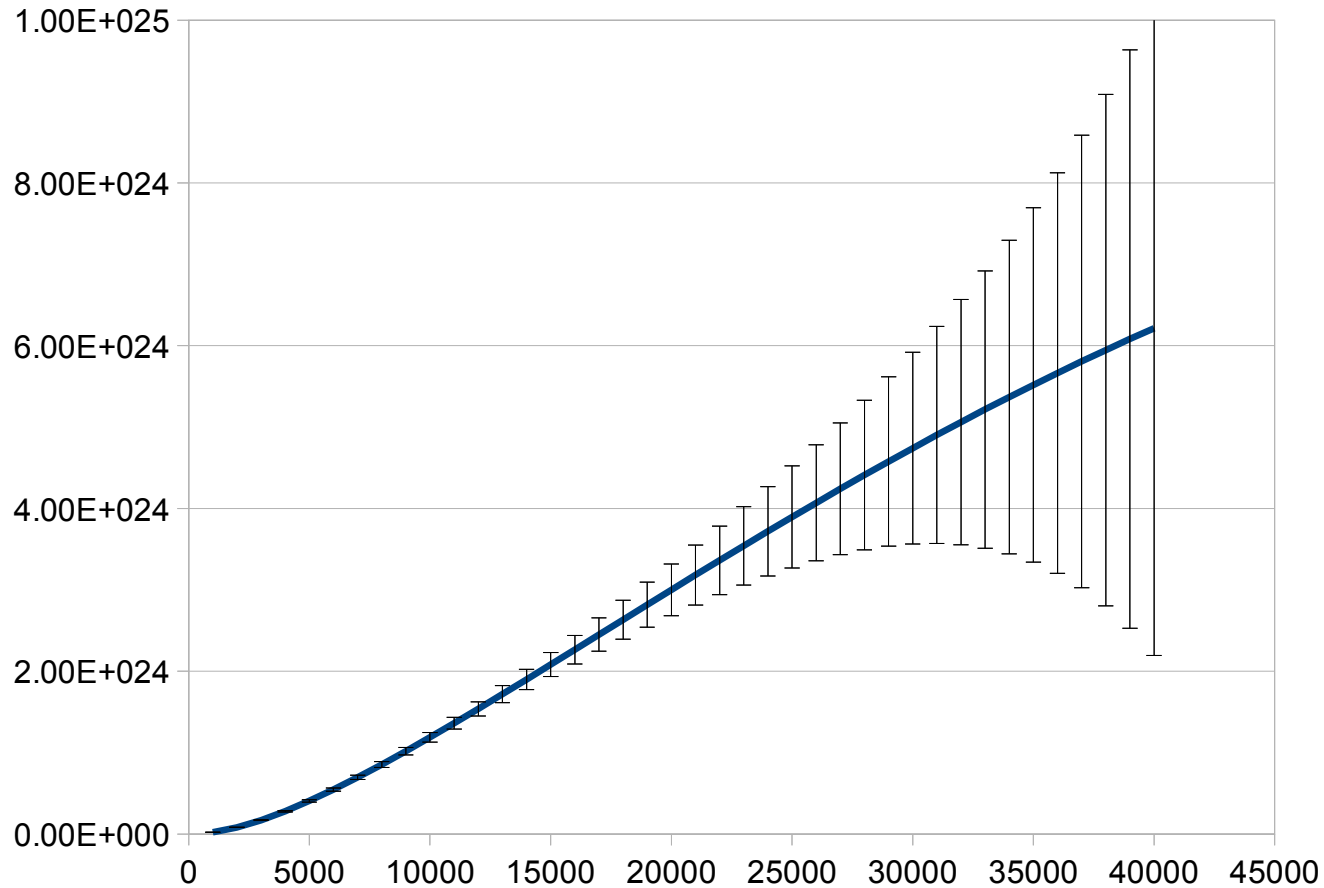
# Simple UOX calculation-Pu239 *Preliminary*



At end  
 $\pm 56\%$

In practice  
C/E =  
 $1.08 \pm 0.07$

# Simple UOX calculation-Pu240 *Preliminary*



At end  
 $\pm 65\%$

In practice  
C/E =  
 $0.99 \pm 0.03$

# Results

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- Large uncertainties occur where production and destruction both important or where parent has large uncertainty
- Adding production and destruction terms uncertainties in quadrature can lead to larger uncertainty terms than the change in number density
- These estimates of uncertainties for Pu239 and Pu240 are much larger than experimental C/E studies have shown
- This over estimation is probably due to correlations between cross-section uncertainties measured.

# Conclusions

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- Decay heat is reasonably well predicted by current codes and data where validation exists. Although improvements still possible.
- This validation can be extended using the available PIE data on individual nuclides, but these typically give larger uncertainties.
- Error propagation could be used to estimate uncertainties on decay heat (and number densities) but would require understanding of the correlations/covariances between evaluated cross-sections, fission yields and decay data.
- It is hoped that such covariance analysis will be part of the proposed ANDES EC project.