

Some thoughts on Fission Yield Data in Estimating Reactor Core Radionuclide Activities (for anti-neutrino estimation)

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Summary



- Introduction
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 - Evaluation
 - Measurements and their analysis
 - Models
 - Adjustment
 - Cumulative and independent yield uncertainty estimation
- Error propagation in nuclear codes
 - "Total Monte Carlo"
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- Some FISPIN calculations of anti-neutrinos in a Pressurised Water Reactor

Introduction



 Anti-Neutrino production in reactors is a decay process and thus is dependent upon the summation over all radionuclides of the decay rate present multiplied by the probability of (anti-) neutrino production.

$$N_{\bar{\nu_e}} = \sum_i \lambda_i N_i * N_{\bar{\nu_e}} \text{ per decay of } i$$

- This probability is equal to the β^+ (or β^-) probability.
- This requires the number densities or activities of the radionuclides present in the core





 The production and destruction of all nuclides are governed by the standard equations:



 Depends on flux, cross-sections, fission yields, decay constants and branching ratios decay paths.



What is nuclear data "evaluation"

- Can be described as the processing of giving numeric value to a quantity
 but
 - Has to be consistent with measurements and constraints of physical laws, at least within currently accepted knowledge.
 - Has to be consistent with semi-empirical or theoretical models.
 - Has to be reported/distributed in a form that all can use easily (e.g. ENDF format)



History of UK evaluations

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1960-1981 Crouch
Atomic and Nuclear Data Tables (1977)
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1981-1987 James and Banai UKFY1 / JEF1 (1986)

1988-1995 Mills, James and Weaver UKFY2 / JEF-2.2 (1993)

1995-present Mills UKFY3.x series - JEFF-3.1.1 (UKFY3.6A)

(UKFY4 photon, neutron and charged particle induced energy dependent or spontaneous fission using Wahl code).



Definitions

The independent yield y(A,Z,I) is the number of atoms of (A,Z,I) produced directly from one fission, but after the emission of prompt neutrons (but before any radioactive decay and hence the emission of delayed neutrons). It can be written as the product of 3 factors:

$$y(A, Z, I) = Y(A) \times f(A, Z) \times R(A, Z, I)$$

where the sum yield or mass yield Y(A) is the total of the independent yields (before delayed neutron emission) of all fission products of mass number A; f(A,Z) is the fractional independent yield of all isomers of (A,Z); and R(A,Z,I), the isomeric yield ratio, is the fraction of (A,Z) produced directly as isomer I.



Independent yields

- Direct probability of forming a nuclide after prompt neutron emission but before decay
- Needed for general inventory calculations
- Difficult to measure, especially for short half-lives or where corrections for major contributions from other nuclides' decay need to be considered.
- In JEFF fits to Wahl Zp model are used to fill extrapolate from small number of measurements to complete set and then adjusted for physical constraints.
- Large uncertainties on values.



Definitions

The **cumulative yield** c(A,Z,I) of nuclide (A,Z,I) is the total number of atoms of that nuclide produced over all time after one fission. If the nuclide is stable the cumulative yield is the total number of atoms of that nuclide remaining per fission after all precursor decays (ignoring the effects of other nuclear reactions e.g. neutron capture). However, for a radioactive nuclide for which this is not the case, some atoms will have decayed before all have been produced.

An equivalent definition that is more useful is the following: immediately at the end of an "infinite" irradiation at the rate of 1 fission per second, c(A,Z,I) is the rate of decay of (A,Z,I) if that nuclide is radioactive, or its rate of production if it is stable.

The chain yield Ch(A) is equal to the sum of all stable or long-lived cumulative yields for a given mass chain. It should be noted that the chain yield, Ch(A), and the sum or mass yield, Y(A), for a mass chain A may differ by a few per cent because the former applies after, and the latter before, delayed neutron emission.



Cumulative yields

- Probability of forming a nuclide directly after prompt neutron emission or by decay, or after a "long" irradiation at 1 fission per second = decay rate (production=destruction)
- Not used in inventory calculations
- Easier to measure and many more measurements
- In JEFF independent yields and decay data used to calculate and then merged with experimental data and their uncertainties using an adjustment process and a "backward-forward" error calculation.
- Smaller uncertainties on values with expt. data

Fission Yield Evaluation



Maximum fraction	of fission ra	Spontaneous	
>10%	1-10%	0.1-1%	fission
Nuclides: 5	2	12	3
233U* TFH	²⁴⁰ Pu* F	²³² Th* FH	²⁵² Cf Sp
235U* TFH	²⁴⁵ Cm TF	²³⁴ U F	²⁴² Cm Sp
²³⁸ U* FH		²³⁶ U F	244Cm Sp
²³⁹ Pu* TF		²³⁷ Np TF	
²⁴¹ Pu* TF		²³⁸ Np TF	
		²³⁸ Pu TF	
		²⁴² Pu F	
		²⁴¹ Am TF	
		^{242m} Am TF	
		²⁴³ Am TF	
		²⁴³ Cm TF	
		²⁴⁴ Cm TF	

- Nuclides in UKFY1 and previous UK libraries [5,7].
- T Thermal fission.
- F Fast fission.
- H 14 MeV fission.
- Sp Spontaneous fission.



Data Analysis

- UKFY3 based upon experimental data from >2000 papers, reports etc.
- Measurements analyzed to given "best estimates"

of each measured yield

Table 1: Number of data items in the UKFY3 experimental database

Absolute	ratio	ratio of ratio	Total		
measurements	measurements	measurements			
11887	1352	1471	14710		

Mass distribution







Historically empirical fitting of Gaussian distributions have been used to model chain yields. Work in the 1960s showed that the best results were obtained using five Gaussians. Due to physical constaints there were only 7 free parameters to fit.

$$\begin{split} \mathsf{Y}(\mathsf{A}) &= \frac{\mathsf{N}_{1}}{\sigma_{1}\sqrt{2\pi}} \begin{bmatrix} \mathsf{e}^{-\left(\frac{(\mathsf{A}-\bar{\mathsf{A}}-\mathsf{D}_{1})^{2}}{2\sigma_{1}^{2}}\right)} + \mathsf{e}^{-\left(\frac{(\mathsf{A}-\bar{\mathsf{A}}+\mathsf{D}_{1})^{2}}{2\sigma_{1}^{2}}\right)} \\ &+ \frac{\mathsf{N}_{2}}{\sigma_{2}\sqrt{2\pi}} \begin{bmatrix} \mathsf{e}^{-\left(\frac{(\mathsf{A}-\bar{\mathsf{A}}-\mathsf{D}_{2})^{2}}{2\sigma_{2}^{2}}\right)} + \mathsf{e}^{-\left(\frac{(\mathsf{A}-\bar{\mathsf{A}}+\mathsf{D}_{2})^{2}}{2\sigma_{2}^{2}}\right)} \end{bmatrix} & \text{with } \mathsf{N}_{3} = 2(1-\mathsf{N}_{1}-\mathsf{N}_{2}). \\ &+ \frac{\mathsf{N}_{3}}{\sigma_{3}\sqrt{2\pi}} \quad \mathsf{e}^{-\left(\frac{(\mathsf{A}-\bar{\mathsf{A}})^{2}}{2\sigma_{3}^{2}}\right)} \end{split}$$



The independent yield is calculated as the integral of a normal distribution

$$\mathsf{FI}(\mathsf{A},\mathsf{Z}) = \frac{1}{2}\mathsf{F}(\mathsf{A},\mathsf{Z})\mathsf{N}(\mathsf{A})(\operatorname{erf}(\mathsf{V}) - \operatorname{erf}(\mathsf{W}))$$

where

$$V = \frac{Z(A) - Z_{p}(A) + 0.5}{\sigma_{z}(A)\sqrt{2}} \qquad W = \frac{Z(A) - Z_{p}(A) - 0.5}{\sigma_{z}(A)\sqrt{2}}$$

F(A,Z) describes the odd-even effect and N(A) is a normalization constant

Charge distribution (Wahl Zp)



Z_p(A) is the most probable charge for mass A

This is calculated as the "unchanged charge distribution" corrected for prompt neutron emission with a term describing the variation of the charge offset ΔZ In the heavy mass peak this is:

$$Z_{p}(A_{H}) = A'_{H}\frac{Z_{f}}{A_{f}} + \Delta Z(A'_{H})$$

In the light mass peak, by conservation of mass and charge

$$Z_p(A_L) = A'_L \frac{Z_f}{A_f} + \Delta Z(A'_{Hc}) \qquad A'_{Hc} = A_f - A'_L$$

The mass after prompt neutron emission is A' and the mass before is A. Thus $A^{\star} = A - \nu(A)$

Charge distribution (Wahl Zp)



 v(A) can be estimated using chain yields by the method of Terrell.



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Charge distribution (Wahl Zp)



The odd-even effect, F(A,Z) is defined by two parameters
 F_Z and F_n.

Where F(A,Z) is given by:

F(A,Z)	proton number Z	neutron number N
$F_Z F_n$	even	even
Ē _Z Ēn	even	odd
Ēn Ēz	odd	even
$\frac{1}{\overline{F}_{Z}\overline{F}_{n}}$	odd	odd



- There exists 388 nuclides with two long-lived isomeric states and 21 with three or more.
- Only small number of measurements.
- The main predictive model available is that of Madland and England that assumed the fragments with a spin near a long-lived isomer would feed that isomer. Model used in UKFY2.
- Rudstam proposed a modification that included energetic feasibility. Results used in UKFY3.





- The emission of protons, deuterons, tritons, alpha particles and other light fragments up to 30 amu have been observed from fission.
- The most common emission is an alpha particle followed by tritons.
- The light charged particles are produced between the two heavy fragments very close in time to the scission of the nucleus.
- In UKFY2 empirical relationships were used to fill gaps, but in UKFY3 improved model results published by Serot et al at ND2004 were used.

Adjustment to physical constraints



$$\cdot \quad \sum_{A} Y(A) = 2.0$$

$$\sum_{A > \frac{A_{f}}{2}} Y(A) = 1$$

• From conservation of mass $\sum_{A} A Y(A) = A_{f} - \overline{v_{\rho}} - A_{LCP}$ • From conservation of charge $\sum_{ZA} Z f(A, Z) Y(A) = Z_f - Z_{LCP}$

and as
$$Z_1 + Z_2 = Z_f$$
 then

$$\sum_A f(A,Z)Y(A) = \sum_A f(A,Z_f-Z)Y(A)$$

for all $Z < \frac{Z_f}{2}$





- Spent Fuel Assay (typically 1 10 years)
- Spent Fuel Decay heat (typically 1 30 years)
- Pulse irradiation of pure fissile species beta/gamma decay heat (typically 1-100000 seconds)
- Delayed neutron emission (typically 1-60 seconds)

Reactor	C/E values	C/E values
	(JEF-2.2)	(JEFF-3.1.1)
Point Beach	0.993 ± 0.016	0.983 ± 0.016
Turkey Point	1.040 ± 0.025	1.026 ± 0.030
Ringhals 2	1.039 ± 0.013	1.025 ± 0.014
Ringhals 3	1.034 ± 0.011	1.021 ± 0.012

Summary of PWR spent fuel decay heatnew validation results.



 Given the individual decay branches for all nuclides in the decay paths from one nuclide to a distant daughter it is possible to calculate the fraction of j that decays to i

$$Q_{j,i} = \sum_{allpaths} \left(\prod_{eachj \to i} B_{j,j+1} B_{j+1,j+2} \dots B_{i-1,i} \right)$$

• If $Q_{i,i}$ is defined as 1 and $Q_{k,i} = 0$ (where k does not decay to i), Thus any cumulative yield can be calculated from the independent yield.

$$Y_i^c = \sum_j Y_j^i Q_{j,i}$$



 As this is a Q weighted sum of Yⁱ then the variance of the result is given by

$$var(Y_i^c) = var(\sum_j Y_j^i Q_{j,i})$$
$$= \sum_j var(Y_j^i) Q_{j,i}^2$$
$$+ 2\sum_j \sum_k Q_{j,i} Q_{k,i} cov(Y_j^i, Y_k^i)$$

 If all terms expect the covariance is known, these can be calculated.



"Forward-Backward" technique

• C is weighted sum of independent yields

C(Z) = I(Z) + I(Z-1)*Q(z-1,z) + I(z-2)*Q(z-2,z) + ...

C(Z+1) = I(Z+1) + Q(z,z+1) * (I(Z) + I(Z-1)) + Q(z-1,z) + I(z-2) + Q(z-2,z) + ...)

C(Z+1) = I(Z+1) + C(Z) * Q(z,z+1)



"Forward-Backward" technique

```
C(Z) = (C(Z+1) - I(Z+1)) / Q(z,z+1)
```

- Equation is now a function of only one cumulative yield, one independent yield and decay data.
- If you know C(Z+1) and its error from measurement and have an estimate of its independent yield and uncertainty can determine uncertainty of C(Z).
- In JEFF-3.1.1, if C(Z) has been adjusted by x to fit independent yield constraints then uncertainty is increased by x.



• What does the Q matrix look like

$Q_{i,j}$	⁸⁵ Ge	⁸⁵ As	^{86}Ga	⁸⁶ Ge	⁸⁶ As	^{85}Se	⁸⁵ Br	^{85m} Kr	^{85}Kr	⁸⁵ Rb
⁸⁵ Ge	1.00	0.85				0.67	0.67	0.67	0.14	0.67
^{85}As		1.00				0.78	0.78	0.78	0.17	0.78
^{86}Ga			1.00	1.00	1.00	0.33	0.33	0.33	0.07	0.33
^{86}Ge				1.00	1.00	0.33	0.33	0.33	0.07	0.33
^{86}As					1.00	0.33	0.33	0.33	0.07	0.33
^{85}Se						1.00	1.00	1.00	0.22	1.00
^{85}Br							1.00	1.00	0.22	1.00
^{85m}Kr								1.00	0.21	1.00
^{85}Kr									1.00	1.00
^{85}Rb										1.00



• The following shows the JEFF-3.1.1 file data and a calculation of Yc and its uncertainty without the covariance terms.

Quantities	^{85}Ge	⁸⁵ As	^{86}Ga	^{86}Ge	^{86}As	^{85}Se	^{85}Br	^{85m}Kr	^{85}Kr	^{85}Rb
JEFF-3.1.1 Y^i	2.44e-5	1.41e-3	1.82e-6	2.85e-6	4.42e-4	9.58e-3	2.19e-3	1.12e-5	4.85e-5	3.30e-8
JEFF-3.1.1 Y^i 1FPY	9.08e-6	4.69e-4	6.65e-11	1.04e-6	1.59e-4	9.47e-4	7.14e-4	4.18e-6	1.81e-5	1.23e-8
$JEFF-3.1.1 Y^{c}$	2.44e-5	1.43e-3	1.82e-10	2.85e-6	4.45e-4	1.08e-2	1.30e-2	1.30e-2	2.86e-3	1.31e-2
JEFF-3.1.1 Y^c 1sd	9.06e-6	4.19e-4	6.66e-11	1.04e-6	1.55e-4	2.10e-4	1.19e-4	1.19e-4	2.10e-4	1.19e-4
Calculated Y^c	2.44e-5	1.43e-3	1.82e-10	2.85e-6	4.45e-4	1.08e-2	1.30e-2	1.30e-2	2.86e-3	1.31e-2
Calculated Y^c 1sd	9.08e-6	4.69e-4	6.65e-11	1.04e-6	1.59e-4	1.02e-3	1.24e-3	1.24e-3	2.68e-4	1.24e-3
Y^c ratio file/calc	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
Y^c 1sd file/calc	0.998	0.893	1.000	1.000	0.978	0.206	0.096	0.096	0.785	0.095

• The results show that without the covariance terms the yields are over-predicted.



- Monte Carlo method is a well understood technique where random sampling of variables is used to solve a mathematical or physical problem.
- "Total Monte Carlo" is where you randomly adjust the parameters going into <u>the evaluation</u> to produce randomised data sets each of which can then be run through a problem to give an output probability distribution on a parameter based upon the entire set of randomised nuclear data that because the evaluation ensure consistency will agree with physical constrains.
 - e.g. TENDL



- Consider a mass of plutonium containing an initial activity of ²⁴¹Pu and ²⁴¹Am. How do these vary with time and what are their uncertainties?
- Given the decay constants are $4.8134 \times 10^{-2} \pm 3.342 \times 10^{-4}$ per year and $1.6019 \times 10^{-3} \pm 1.852 \times 10^{-6}$ per year respectively.
- The "best estimate" result can easily be calculated.
- If we sample the decay constants from different probability distributions with the required mean and standard deviation, run each for 10000 times and then analyse the results we get

Probability Distribution functions





Result





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²⁴¹Am

3933

28

3933

28

3933

27

±

±

±

Plot of 255 ²⁴¹Pu decay curves from normal distribution calcs







- In nuclear data work it is often necessary to consider larger uncertainties, even orders of magnitude.
- Repeating the calculations using 50 percent uncertainties on the decay constants, we get activities at 200 years for

 241 Pu activities of 4.62x10⁵±3.42x10⁷, 560±1762 and 2595±7094 (should be 10.1) and ²⁴¹Am activities of 18946±727627, 4683±2888 and 5522±5221 (should be 3933)

Larger uncertainties



- Mean and standard deviations of $^{241}\text{Pu}\;\lambda$ are
 - Normal 0.048224 ± 0.02405
 - Linear 0.048175 ± 0.02411
 - LogNormal 0.048210 ± 0.02403
- Compared to input 0.048134 looks okay!
- Examining PDFs and results ...

Probability Distribution functions



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Plot of 255 ²⁴¹Pu decay curves from normal distribution calcs





Decay example Conclusions

- This shows the method fails with high uncertainties, resulting from their physical interpretation.
- What do these large uncertainties mean?
- An uncertainty of, say, > 35 percent has little meaning without specifying its distribution and any constraints on the value from the expt. analysis, theoretical model or evaluation process.
 - For example λ must be greater than zero.
- Without these it may only be possible to quote a "best estimate" result.





- TENDL-2010 included FPY and decay data files with up to 1000 randomly perturbed data libraries for used in uncertainty propagation.
- Using these FPY libraries, the UK spent fuel inventory code FISPIN was used to calculate decay heat from a fission pulse with each library.
- The unperturbed JEFF-3.1.1 decay data library was used and thus the uncertainties from energy release per decay and half-lives are not considered. Although some published Algora/Tain TAGS data was included extending JEFF-3.1.1.
- The results were compared Tobias (1989).

²³⁹Pu total energy release





²³⁵U total energy release





²³⁹Pu γ-ray energy release





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²³⁵U γ-ray energy release





²³⁹Pu β -particle energy release





235 U β -particle energy release



0.9

8.0

0.7

0.6

0.5

0.4

0.3

0.2

0.1

Energy release rate per fission x time (MeV/s / fission).s

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- As expected the TENDL results show larger uncertainties at shorter times when more shortlived fission products contribute. Note scaling by multiplying by time, reduces value and uncertainties below 1 second.
- The results show the expected under-prediction for ²³⁵U gamma-ray energy release reported by other workers. As the corresponding beta particle energy release appears centred about the Tobias data it appears that this is more about fission yields rather than only a decay data effect.



- Consider a typical modern PWR reactor containing 75.14 metric tonnes of uranium in a UO_2 fuel with a 4.5% enrichment.
- Consider a mean rating of 45.833 MW/t (i.e. a total power of 3444 MW thermal) with fuel residing in the core for four 300 day cycles with 30 day gaps for refuelling and maintenance what neutrino source term would we expect.
- In a real core there will be variation both axially and radially in the core e.g. at the end of the assemblies where the power could considerably less.
- The following considers the mean power and to show effects 20% of this but with the same residency time.



- Using the FISGUI (FISPIN Graphical User Interface) calculations of were made every 30 days during irradiation and shutdown, after the irradiation the fuel was considered as removed and cooled to 1, 30, 50, 100 and 365.25 days.
- The activities were then multiplied by the probability of β^- emission and summed to get the neutrino emission (i.e. no weighting by detection probability).



Anti-neutrino emission rate (per second) per tonne of fuel against time in days.





Fractional anti-neutrino contribution 30 days into cycle

	First cycle						Fourth cyc	le		
#	Nuclide	half-life (d)	fraction	cumulative		#	Nuclide	half-life (d)	fraction	cumulative
1	U239	0.0163	0.07534	0.07534		1	U239	0.0163	0.0984	0.0984
2	NP239	2.3550	0.07510	0.15044		2	NP239	2.3550	0.0980	0.1964
3	1134	0.0365	0.01236	0.16280		<u>3</u>	134	0.0365	0.0109	0.2072
4	CS138	0.0224	0.01074	0.17354	\land	4	1133	0.8667	0.0098	0.2170
5	TE134	0.0291	0.01072	0.18426		5	XE133	5.2450	0.0095	0.2265
6	1133	0.8667	0.01068	0.19494		6	1135	0.2754	0.0092	0.2358
7	XE133	5.2450	0.01040	0.20534		7	CS138	0.0224	0.0092	0.2449
8	BA139	0.0577	0.01034	0.21568		8	MO99	2.7500	0.0090	0.2539
9	Y95	0.0072	0.01031	0.22599		9	XE137	0.0027	0.0089	0.2627
10	CS139	0.0064	0.01026	0.23625		10	NB100	0.0000	0.0085	0.2713
11	Y94	0.0133	0.01024	0.24649		11	BA139	0.0577	0.0085	0.2798
12	XE138	0.0098	0.01024	0.25673		12	TE134	0.0291	0.0085	0.2883
13	I135	0.2754	0.01020	0.26692		13	TC101	0.0099	0.0085	0.2968
14	Y93	0.4208	0.00998	0.27690		14	MO101	0.0101	0.0085	0.3053
15	MO99	2.7500	0.00996	0.28686		15	NB98	0.0000	0.0083	0.3136
	241 nuclide	es contribut	te 99.5%				292 nuclid	es contribut	te 99.5%	
	1							1	I	

50



Fractional anti-neutrino contribution after irradiation

1 Day					-	1)	ear cooling			
#	Nuclide	half-life (d)	fraction	cumulative	-	#	Nuclide	half-life (d)	fraction	cumulative
1	NP239	2.355	0.33	0.33	-	1	PR144	0.012	0.19	0.19
2	XE133	5.245	0.04	0.37		2	CE144	284,896	0.19	0.38
3	LA140	1.678	0.03	0.40		3	RU106	368,200	0.13	0.51
_ 4	RU103	39.350	0.03	0.44		4	RH106	0 000	0.13	0.64
5	BA140	12.740	0.03	0.47	/ / / \ \	5	CS134	753 146	0.07	0.71
6	CE141	32.500	0.03	0.50		6	PU2/1	5259 594	0.06	0.76
7	NB95	35.150	0.03	0.53		Ŋ	CS137	40057 408	0.00	0.70
8	ZR95	63.980	0.03	0.56		-	03137	10957.490	0.05	0.02
9	PR143	13.580	0.03	0.59		ŏ	PM147	958.051	0.05	0.87
10	MO99	2,750	0.03	0.62		9	Y90	2.667	0.03	0.90
11	CF144	284 896	0.02	0.64		10	SR90	10636.074	0.03	0.94
12	PR144	0.012	0.02	0.66	-	11	NB95	35.150	0.02	0.96
13	1132	0.096	0.02	0.69		12	ZR95	63.980	0.01	0.97
14	TE132	3.258	0.02	0.71		13	EU154	3141.146	0.01	0.98
15	Y91	58.510	0.02	0.73	-	14	SB125	996.447	0.01	0.98
16	1131	8.040	0.02	0.75	-	15	Y91	58.510	0.01	0.99
17	1133	0.867	0.02	0.77		16	KR85	3915.474	0.00	0.99
18	CE143	1.375	0.02	0.79		17	EU155	1810.417	0.00	0.99
19	RH105	1.473	0.02	0.80		18	SR89	50.500	0.00	1.00



- Anti-neutrino emission is approximately proportional to reactor power.
- Varies between 1.96E20 and 2.18E20 neutrinos per GW for a single assembly
- Small variation of value during irradiation $\sim 10\%$.
- Assuming all fuel reaches the same final irradiation in 4 cycles an equilibrium core will have a starting value of 20.65 GWd/t and finishing value of 34.75 GWd/t (mid-point 27.5 GWd/t)
- Then expect neutrino emission to be nearer to 2.07E20 per GW with ~3% variation during cycle.
- In a real core expect variation to be larger.