



Some of the work performed at BNL in the last 3.75 years

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ENDF/B Decay Data Sub-library

Decay data for all known nuclides, 3,821 materials, that is, stable and long-lived ground state and isomeric levels.

Mostly based on the Evaluated Nuclear Structure Data File (ENSDF).

New JSON format coming soon!

7=28

7=20

Atomic data using BRICC, LOGFT codes to calculate vacancies and EADL data to propagate vacancies out.

N=126

Incorporates theoretical (CGM code, T. Kawano *et al.*) gamma, electron, antineutrino and neutron data for neutron-rich nuclides with non-existent or incomplete decay data.



N=50

7 = 50

7=82

N=82



ENDF/B Contains TAGS data for 55 materials:

⁸⁶Br (ORNL), ^{87,88}Br (Valencia), ^{90,90m,91,93}Rb (INL), ⁹⁴Rb (Valencia), ⁹³Sr (Greenwood), ⁹⁵Y (INL), ¹⁰¹Nb (Valencia), ^{103,104}Nbm (MSU), ¹⁰⁵Mo (Valencia), ^{102,104,105,106,107}Tc (Valencia), ^{140,141}Cs (INL),¹⁴²Cs (ORNL), ^{141,142,143,144,145}Ba (INL), ^{142,143,144,145}La (INL), ^{145,146,147,148}Ce (INL), ^{146,147,148,148m,149,151}Pr (INL), ^{149,151,153,154,155}Nd (INL), ^{152,153,154,155,156,157}Pm (INL), ^{157,158}Sm (INL), ¹⁵⁸Eu I(INL).

IB adjusted to match the electron spectra measured by Tengblad *et al.* for:

⁸²As, ⁸⁹Br, ⁹⁰Br, ^{95,96}Rb, ^{98,99}Y, ¹³⁴Sb, ¹³⁸I

ENDF/B available from the NNDC's GitLab server



Consistency Issues

In ENSDF, absolute gamma + CE, $I\gamma$, and beta intensities, $I\beta$ -, are related by the intensity balance at each level.

In ENDF/B we use TAGS data for beta intensities and ELP, <Ee->, and EEM, <E γ >.

The use of TAGS data in ENSDF and ENDF/B breaks the intensity balance, creating inconsistencies, that must be documented to alert the user. ENSDF evaluators often give the TAGS I β - as comments

If the ENSDF data agrees *within 10%* with the TAGS data, then we use ENSDF to avoid inconsistencies.

There are also inconsistencies if we use ELP, EEM and I β s from TAGS and theoretical gamma, & neutron if present, spectra from CGM.



TAGS data effect on ²³⁵U electron spectrum

Ratio of ²³⁵U electron spectra measured at ILL by Schreckenbach *et al.* to two different summation calculations

Below 5 MeV, the use of JEFF-3.3 decay data overpredicts the spectrum because it doesn't include beta intensities from TAGS.

For higher energies, the underprediction is mainly from the lack of theoretical spectra in JEFF-3.3.

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New Stuff!



ORNL delayed gamma and electron data

- A couple of NNDC library bookshelves collapsed in May 2020.
- Among them we found three very valuable reports with delayed electron and gamma spectrum following the thermal fission of ²³⁵U and ^{239,241}Pu.



Only one report available online, which can't be searched by content.



We knew of this data because it is the only decay heat measurement at times shorter than 20 seconds, and that for larger times agree quite well with the Yayoi measurements.





Reconsideration of the Theoretical Supplementation of Decay Data in Fission Product Decay Heat Summation Calculations N. Hagura, T. Yoshida, T. Tachibana, Journal of Nuclear Science and Technology, 43:5, 497 (2012) IAEA 2023 TM Antineutrinos – A.A. Sonzogni *et al.*

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Pioneer ²³⁵U antineutrino spectrum

We later learned that the antineutrino spectrum derived by Dickens from this data, published in 1981, agrees quite well with that from Huber published in 2011.

Cited by P. Vogel 1981 seminal summation work, but not cited by anyone else.





ORNL irradiations

Nucleus	Irradiation time	Delay time	Counting time	Number of measurements
²³⁵ U	1 s	1.7 s	110 s	14
²³⁹ Pu	1 s	1.7 s	130 s	15
²⁴¹ Pu	1 s	1.7 s	130 s	15
²³⁵ U	10 s	10.7 s	795 s	14
²³⁹ Pu	5 s	17.7 s	1,198 s	15
²⁴¹ Pu	5 s	17.7 s	1,198 s	15
²³⁵ U	100 s	69.7	13,500 s	15
²³⁹ Pu	100 s	250 s	13,950 s	13
²⁴¹ Pu	50 s	195 s	13,975 s	14



ORNL delayed gamma and electron data

We scanned and digitized 260 tables in them.

The three reports were also converted to PDF with a high-quality scanner.



SPECTRUM OF BETA 1-SEC THEBRAL- VEUTRON	RAYS FOLLOWING A IRRADIATION OF 235-0
START COUNT 1.7 SEC AFT COUNT FOR	TE IND OF IBRADIATION
E(BITA) T(BITA) DELTA(T)	E (BETA) T (BPTA) DELTA (T)
0, 170 6 1655-02 4 7355 02	HET BETAS/MEY/PISSION
0, 190 6 1378-02 1.7258-02	2.360 2.876E-02 2.376E-03
0.210 6.5262-02 1.5032-02	2.440 2.9798-02 2.2258-03
0.230 6.3068-02 1.4168-02	2. 520 2.7522-02 2.2362-03
0.250 5.0388-02 1.4288-02	2.680 2.5416-02 1.9926-03
0.275 3.4832-02 1.3512-02	2.760 2.4556-02 2.0286-03
0.305 4.4658-02 1.2618-02	2.840 2 1102-02 1.7902-03
0.335 5.053E-02 1.200E-02	2,920 2,285 8-03 1 7088-03
0.365 J.8682-02 1.181E-02	3,000 2,1612-02 1,7981-03
0.395 4.900E-02 1.040E-02	3.080 1.9797-02 1 6052-03
0.425 5.634E-02 8.0802-03	3,160 1,9122-02 1,6612-03
0.455 4.4872-02 6.4732-03	3,250 1,7318-02 1 8538-03
0.485 4.1242-02 6.3702-03	3.350 1.5118-02 1.4598-03
0.520 4.157E-02 6.210E-03	3.450 1.4238-02 1.4278-03
0.560 3.8065-02 6.0075-03	3.550 1.3412-02 1.3252-03
0.600 4.655E-02 5.720E-03	3.650 1.194E-02 1.211E-03
0.640 4.6502-02 5.4942-03	3.750 1.124E-02 1.192E-03
0.680 4.1705-02 5.0415-03	3.860 1.0962-02 1.1182-03
0.720 4.5422-02 5.2192-03	3.980 8.0772-03 9.773E-04
0.760 4.7232-02 5.0278-03	4.100 7.076E-03 8.971E-04
0.800 4.5635-02 4.8475-03	4.220 8.038E-03 9.009E-04
0.840 4.4922-02 4.3142-03	4.340 7.549E-03 8.592E-04
0 825 4 1058-02 4 2248-03	4.460 6.097E-03 7.555E-04
0.975 3 0102-02 4 0522 03	4.580 5.240E-03 7.304E-04
1.025 3.9278-02 3.0128-02	4.700 4.450z-03 6.177z-04
1.075 3.9168-02 3.7928-03	4.820 3.616E-03 6.158E-04
1,125 4, 1912-02 3, 7728-03	4.940 3.2988-03 5.2908-04
1. 175 4. 2278-02 1. 5968-03	5.070 3.512E-03 5.573E-04
1.225 3.8258-02 3.4508-03	5 350 3.11/E-03 5.103E-00
1.275 4.0062-02 3.4432-03	5.490 1 1865-03 3.9412-04
1.325 4.4342-02 3.1592-03	5.630 8.6132-00 3.3312-00
1.375 4.259E-02 3.204E-03	5 770 1 2068-03 1 0038-04
1.430 3.922E-02 3.117E-03	5.910 1.6618-03 1.7058 04
1.490 4.012E-02 3.046E-03	6.050 1.5638-03 3 2038 00
1.550 4.006E-02 3.078E-03	6, 190 1, 1798-03 2 9628-04
1.610 3.8992-02 2.9292-03	6.330 8.528P-04 2 858P-04
1.670 4.172E-02 2.833E-03	6.480 5.584E-08 1.968E-08
1.730 4.210E-02 3.054E-03	6.640 3.5962-04 1.5562-04
1.790 3.591E-02 2.930E-03	6.800 3.175E-04 1.506E-04
1.850 3.206E-02 2.776E-03	6.960 3.0002-04 1.3602-04
1.910 3.434E-02 2.729E-03	7.120 2.2058-04 1.0958-04
1.970 J.6J8E-02 2.796E-03	7.280 1.1638-04 8.7828-05
2.040 3.519E-02 2.687E-03	7.440 4.3448-05 7.6428-05
2 200 3 2302 02 2 5928-03	7.600 1.2472-05 7.2162-05
2,280 2,9898-02 2,4618-03	7.760 6.5462-06 7.5802-05

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SPECTRUM OF GAMMA RAYS FOLLOWING A 1-SEC THERMAL-NEUTRON IRRADIATION OF 235-U

START COUNT 1.7 SEC AFTER EVO OF TRRADIATION COUNT FOR 1 SEC

ELGAMMAI	VEGANMAS DELTACYS	ELGANNAL	YEGANNAL DELTAINE
NEV	GAMMAS/WEV/FISSION	NEV	SAMMA S/MEV/ETSSIDN
0.055	A-6448-02 7.2948-02	1 040	1414 11 11 11 11 11 11
0.065	5.8191-02 2.1715-02	1.0.00	2.4010-02 4.4472-03
0.075	8 9935-02 2 5715-02	1.980	2.3/58-02 4.3908-03
0.015	8.9020-02 2.578E-02	2.020	2.0738-02 4.1756-03
0.005	0.504C=02 2.711E=02	2.060	2.106E-02 4.161E-03
0.045	1.8756-01 3.2288-02	2.100	1.9588-02 3.9348-03
0.105	1.9992-01 3.2938-02	2.140	1.6346-02 3.7316-03
0.115	3.3036-01 3.8856-02	2.180	1.5816-02 3.6416-03
0.125	3.5026-01 3.9486-02	2.220	1.5935-02 3.7325-03
0.135	2.1016-01 3.1946-02	2.260	1.6316-02 1.6305-03
0.145	2.2126-01 3.1858-02	2.300	1.7905-02 1 4005-01
0.155	2-1465-01 3-1905-02	2.140	1 84 35-02 3.0000-03
0.145	1-8615-01 3-1475-02	2 3 80	1.0420-02 3.0300-03
0-177	1-7446-01 2.7875-02	2 . 36	1.0000-02 3.7102-03
0.192	1 5045-01 2 5525-02	61969	1.3708-02 3.3928-03
0.207	1.5040-01 2.5500-02	2. 13	1.4288-02 3.5186-03
0.207	1.4402-01 2.4302-02	2.525	1.614E-02 3.686E-03
0.222	9.373E-02 2.157E-02	2+575	1.4246-02 3.4106-03
0+231	9.042E-02 2.080E-02	2.625	8.989E-03 3.084E-03
0.252	1+302E-01 2.219E-02	2+6.75	1.0198-02 3.0188-03
0.267	1.356E-01 2.269E-02	2.725	1.3706-02 3.4936-03
0.282	1.4536-01 2.2856-02	2.775	7.259E-03 2.879E-03
0.297	1.770E-01 2.422E-02	2.825	7.4105-01 2.8405-03
0.313	L-115E-01 1.994E-02	2.875	1.2545-02 3.1555-03
0.327	7-916E-02 1-719E-02	2.925	9.3745-03 3.8185-03
0.342	9.2285-02 1.7935-02	2.075	# 2176-D1 2 3116-01
0.357	1.0095-01 1.9255-02	8 0.20	1 1 2 2 6 0 2 2 0 1 1 2 0 3
0.372	1.0775-01 1.8505 02	3.000	1.1230-02 2.0038-03
0.342	1 2225-01 1.8546-02	3.040	0.1032-03 2.5422-03
0.507	1.337E-01 1.917E-02	3.150	7.0766-03 2.4166-03
0.402	1.540E-01 1.980E-02	3.210	1.1788-02 2.6388-03
0.417	1.209E-01 1.808E-02	3.270	1.2598-02 2.8428-03
0.432	1.0516-01 1.6706-02	3.330	1.1258-02 2.5678-03
0.447	1.2816-01 1.2276-02	3.390	1.2005-02 2.7365-03
0.462	1.7326-01 1.3408-02	3.450	8.7195-01 2.2795-01
0.477	1.7825-01 1.1905-02	3.510	6.1525-01 1 0205-01
0.692	1-552F-01 1-267E-02	3.573	5 9015-03 1 8785-03
0.507	1-5655-01 1-2078-02	3 6 30	4.9010-03 1.0700-03
0.522	1.8955-01 1.4155-01	3.0.50	4.0502-03 1.005E-03
0. 5.0	1.0 970-01 1.4150-02	3+0.90	1.9846-03 1.5906-03
0.540	2.4176-01 1.5636-02	3+ 750	3.3536-03 1.7046-03
0.500	2.1.446-01 1.4696-02	3.810	4.129E-03 1.875E-03
0.580	1.569E-01 1.277E-02	3.873	2.687E-03 1.739E-03
0.600	1.0100-01 1.2020-02	3.935	2.998E-03 1.785E-03
0.620	1.224E-01 1.104E-02	4.005	2.645E-03 1.671E-03
0.640	8.8618-02 9.4648-03	4.075	2.566E-03 1.435E-03
0.660	7.754E-02 9.320E-03	4.145	5-561E-03 1.777E-03
0.680	6.201E-02 8.234E-03	4.215	A- 347E-03 1.584E-03
0.700	5-975E-02 8-494E-03	4.285	5.120E-03 1 ALBE-01
0.720	5-6205-02 8-0.085-03	4.345	5 5385-33 1 4845 03
0.740	5-855E=07 7 930E=03	4 4 75	7. 7 37E-33 1.070E-05
0.740	4 5585-02 8 5518-03	4.425	8.104E-03 1.529E-03
0.780	7 4485-02 8 7445-03	4. 7. 72	5-277E-05 1+310E-03
0.100	1.0492-02 0.1042-03	4.205	8.540E-04 8.732E-04
0.800	1.1652-01 1.0052-02	4.635	8.829E-04 1.002E-03
0.820	1.382E-01 1.045E-02	4.705	1.284E-03 8.776E-04
0.840	1.0288-01 9.1318-03	4.775	1.2918-03 8.6628-04
0.860	6.998E-02 7.751E-03	4.845	6.989E-04 7.474E-05
0.880	6.356E-02 7.960E-03	4.915	1.546E-04 5.911E-04
0.900	5.671E-02 7.393E-03	4.985	4.240E-04 6.745E-04
0.920	5.635E-02 7.550E-03	5.060	1.217E-03 7.666E-05
0.940	7.137E-02 8.014E-03	5.140	1.589E-03 8.047E-34
0.962	8.2546-02 8.4246-03	5.220	1.1115-03 7.3335-05
0.987	7.2955-02 8.0225-03	5.300	5.972E-06 5.526E-06
1.013	A-109E-02 7-368E-03	5.380	3. 9785-04 5. 8355-04
1.037	5.5385-02 7.4155-03	5.440	A 211E-04 5 4045 AT
1.042	5.8418-02 7 3835-03	5 5 400	4 7345-04 5.4942-04
1.0.00	7 8115-02 8 0855-03	5.540	0.120E-04 3.351E-04
1.000	8 0715-02 8.0050-03	3.020	3.4902-04 4.45EE-04
1.112	0. 011E-02 0.352E-03	5.700	5-4135-04 2-5456-04
1+130	7.4716-02 7.6546-03	5.780	3.8198-04 6.1216-04
1.162	5.264E-02 6.828E-03	5.860	3.3168-04 5.2478-04
1.187	4.898E-02 6.692E-03	5.945	7.5308-05 4.3918-04
1.215	6.253E-02 7.215E-03	5.035 -	-1.195E-04 3.27#E-04
1.245	4.322E-02 7.249E-03	5.125	5.305E-05 3.217E-05
1.275	5.428E-02 6.606E-03	0.215	5.742E-04 4.353E-04
1.305	5.0406-02 6.8336-03	6.305	8.111E-04 4.750F-05
1.335	3.724E-02 5.786E-03	6.395	5.998E-04 3.860E-05
1.365	3.157E-02 5.917E-03	6.4.85	5.228E-04 3.273E-04
1.395	4-118E-02 5-958E-03	6.575	2.4365-04 2.6275-04
1.625	4-050E-02 6-0685-03	5.645	9-048E-05 2 144E-04
1.455	2.0028-02 5.0105-03	4.745	3. 9985-05 2. 0705 21
1.4.85	2.6335-02 5 1235-03	6.853	5 4035-05 2.0142-04
1.515	1 1 245-02 5 0415 03	6.050	1.0030-03 1.0140-04
1.515	3.1222-02 3.0612-03	5.950	0.000E-05 1.753E-04
1.545	2.979E-02 4.909E-03	7.050	7.264E-05 1.697E-04
1.580	3.310E-02 5.226E-03	7.150	5.288E-05 1.613Ec04
1.620	3.200E-02 5.109E-03	7.250	3.127E-05 1.587E-04
1.660	2.5718-02 4.7128-03	7.350	1.7698-05 1.4705-04
1.700	2.474E-02 4.637E-03	7.450	2-533E-05 1-380E-05
L. 740	2.9976-02 4.8418-03	7.550	1.456E-05 1.395E-04
1.780	3-316E-02 5-164E-03	7.650	9-129E-06 1-398E-04
1.820	2.704E-02 4.631E-03	7.750	1.5296-05 1.3355-05
1.860	2.6738-02 4.6675-03	7.850	1.9045-05 1.2785-04
1.900	2.5495-02 4.4815-03		1.1000-03 1.2002-04

We are grateful to SULI interns Zharia Harris, Becket Hill, Bryan Palaguachi and Matthew Seeley, who help digitizing and understanding the data.





Beta-delayed electrons

Measurements are compared with summation calculations, ENDF/B-VIII.1β decay and JEFF-3.3 yields, which highlights the 25 most important contributors.

Good agreement is seen for ²³⁵U, in this case 1-second irradiation, 6.7second waiting period, and 3-second counting interval.

Brookhaven National Laboratory





Gamma spectrum per fission

What percentage of the equilibrium spectrum do these irradiations capture?

- The 1-second irradiation captures about 30-65% of the equilibrium spectrum in the 1 to 8 MeV region.
- The medium and long irradiations capture considerably less, therefore correction is larger, that is, more reliance on the summation method.
- Differences between JEFF-3.3 and ENDF/B-VIII.0 decay data are due to the implementation of TAGS data in the ENDF/B.





Issues

- We don't have the full data set, despite that we searched for it, with the help of David Glasgow from ORNL.
- We can calculate ratio of electron spectra similar to Kopeikin et al, but corrections are needed.
- The uncertainty of the sum spectra are not available, need to fix that.





Issues

- To check and possibly reassure ourselves, we can calculate the ratio of the medium/short irradiation and apply corrections.
- The corrected ratio should be closed to one.
- Uncertainties include summation model uncertainties using a Monte Carlo method.





Results!

- Both summation and derived ORNL values are lower than ILL ones.
- For energies lower than 3.5 MeV the ORNL values are very close to Kopeikin's.
- At 4 MeV, the ORNL values are about half-way between ILL and Kopeikin's.
- Note that for energies higher than 7.5 MeV, ILL and Kopeikin's values unexpectedly drop, which is not supported by summation calculations, a ²³⁵U or ²⁴¹Pu contamination in the ²³⁹Pu target could be the reason.



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Results!

- Both summation and ORNL values are lower than ILL ones.
- Seems to indicate that the ²³⁵U ILL electron spectrum is too high.
- ILL ratio shape agrees with summation for high energies.





Results!

- Both summation and ORNL values are higher than ILL ones.
- Normalization issues in the ²³⁹Pu and ²⁴¹Pu ILL electron spectra are possibly present.
- ILL ratio values at high energies disagree with summation values. ²⁴¹Pu fission products are more neutron rich than ²³⁹Pu ones, therefore they produce more energetic electrons. A contamination in the ²³⁹Pu target could be the reason.





Additionally,

- The modified Kopeikin ²³⁵U and ²³⁸U antineutrino spectra combined with Huber's ²³⁹Pu and ²⁴¹Pu do not agree that well with the Daya Bay spectra
- The overprediction (dip) at 2-5 MeV and underprediction (bump) at 5-7 MeV persist.
- We definitely need newly measured electron spectra to properly account for the antineutrino spectra produced by nuclear reactors.





Daya Bay High Energy Neutrinos









Electron Spectrum (1/MeV Fission)

We are unable to reproduce the Daya Bay high energy segment of the spectrum with our current summation calculations

Antineutrino Spectrum





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What's the maximum antineutrino spectrum a betaminus decaying nucleus can have?

Maximum energy electron and antineutrino production





1.20

We are able to better reproduce by adjusting ^{102g,mY} and ¹⁰⁴Nb

Other important cases are ^{92,93}Br, ⁸⁶As, ¹⁴⁰I, ¹⁴⁴Cs, ¹⁰⁴Y



Fine structure, yes? no? may be? who knows?



Analysis of Daya Bay's arXiv:2102.04614v2 data

Spectrum and its covariance given with 50 keV prompt energy bin.

We will re-bin the data and obtain new covariance matrices using:



 $cov(aX + bY, cW + dV) = ac \ cov(X, W) + ad \ cov(X, V) + bc \ cov(Y, W) + bd \ cov(Y, V)$

The presence of individual fission products (Fine Structure) is revealed using the ratio of adjacent spectrum values:

$$R_i = S_i / S_{i+1},$$

with uncertainty given by:





With σ the covariance matrix.

To better reveal fine structure you need to reduce the spectrum uncertainty and increase the adjacent spectrum points correlation!

Our 2018 analysis





Phys. Rev. C **98**, 014323 (2018) ³³





ותבת בטבט דואו תוונוופענוווטט – ת.ת. טטוובטעווו כו מו.



For this particular case, the sweet spot to reveal fine structure seems to be a 250 keV binning.

Note:

To understand the possible individual fission product effects, we really need the spectrum as function of the antineutrino energy.

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Conclusions

We really need to remeasure the big 4's electron spectra with modern techniques and 25 keV bins.

- We absolutely need TAGS data to predict decay heat and nuclear reactors antineutrino spectrum. There are still approx. 20 nuclides waiting for a measurement.
- Keep in mind that most relevant fission yields have not been measured directly, isomers can be a problem.

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We are grateful to the Daya Bay collaboration for sharing their data, including a full documentation and spectrum covariance matrix.



Backup material









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$$\Delta^{2}S_{stat}(E) = \Delta^{2}S_{stat}(E) + \Delta^{2}S_{sys}(E)$$

$$\Delta S_{stat}(E) = c_{stat}S^{1/2}(E)$$

$$\Delta S_{sys}(E) = c_{sys}S(E)$$

$$\int_{0}^{\infty} \int_{0}^{\infty} 1 \times 10^{5}$$

$$\int_{0}^{\infty} \int_{0}^{\infty} \int_{0}^{\infty} 1 \times 10^{5}$$

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