Analysis of ^{235}U and ^{239,241}Pu delayed electron and gamma spectra measured by J.K. Dickens *et al.*

Z. Harris[1], B. Hill[1], S. Kim[2], A. Mattera[2], E.A. McCutchan[2], B. Palaguachi[1], M. Seeley[1], A.A. Sonzogni[3]

[1] Science Undergraduate Laboratory Internships (SULI)

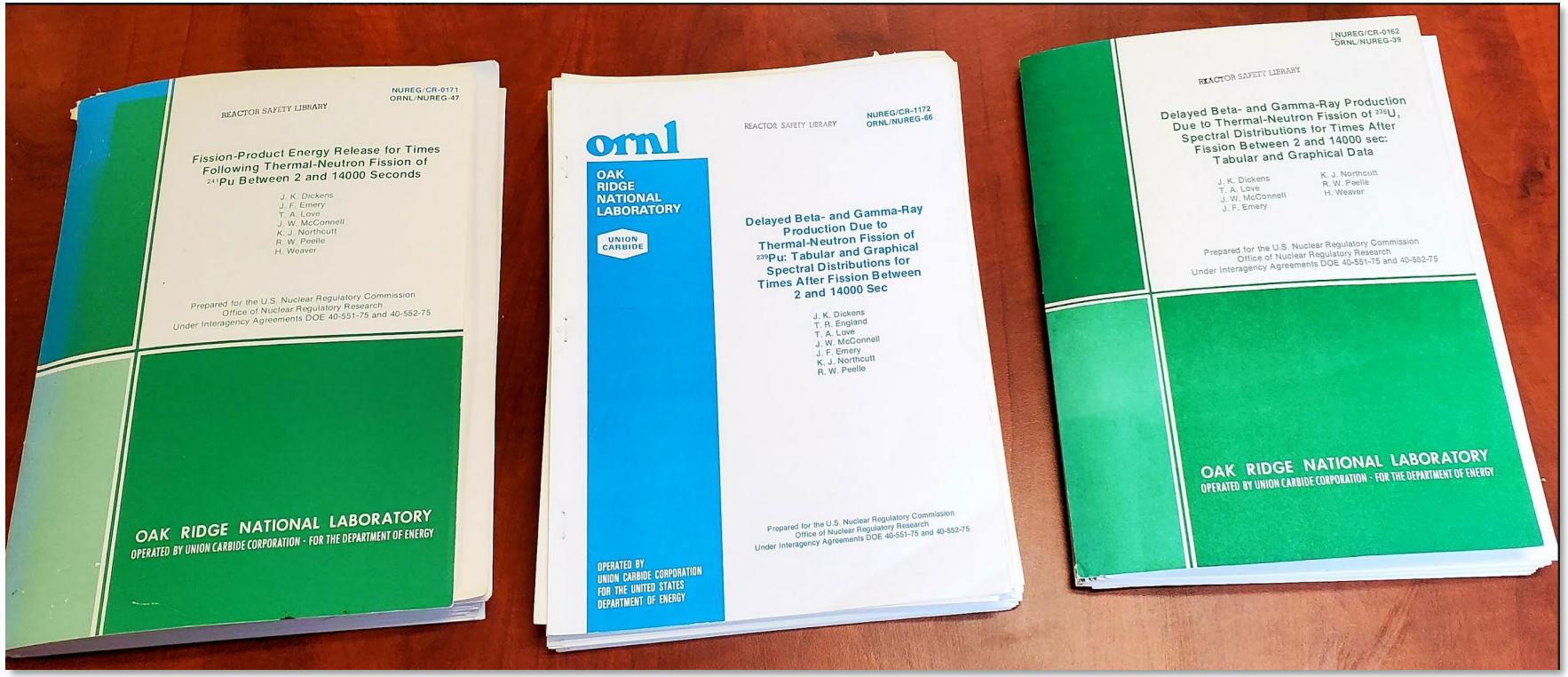
[2] National Nuclear Data Center

[3] Nuclear Science & Technology Department

Work sponsored by the Office of Nuclear Physics, Office of Science of the U.S. Department of Energy under Contract No. DE-AC02-98CH10886, as well as by the U.S. Department of Energy, National Nuclear Security Administration, Office of Defense Nuclear Nonproliferation Research and Development (DNN R&D).

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 ^{235}U and $^{239,241}Pu$.

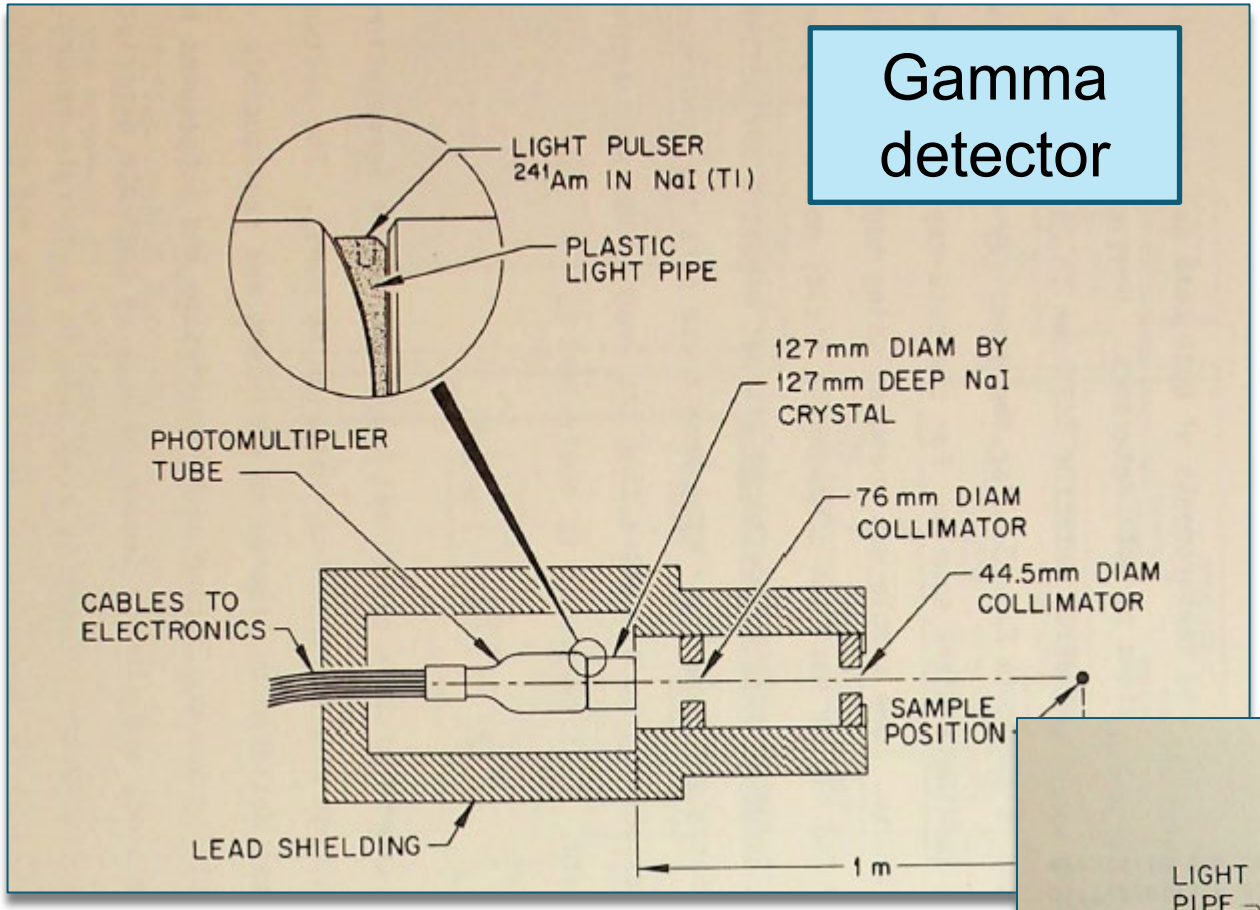


Only one report available online, which can't be searched by content.

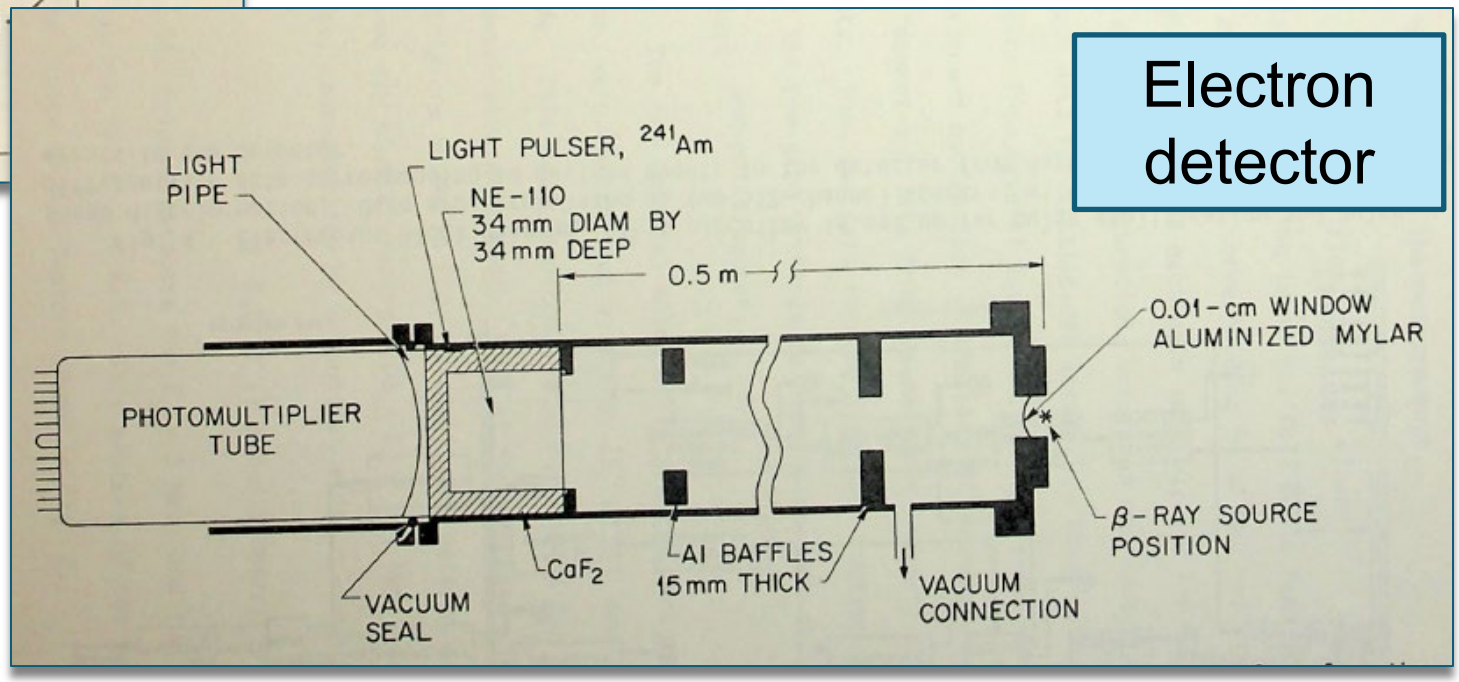
Brookhaven National Laboratory

ORNL detectors

Gamma detector

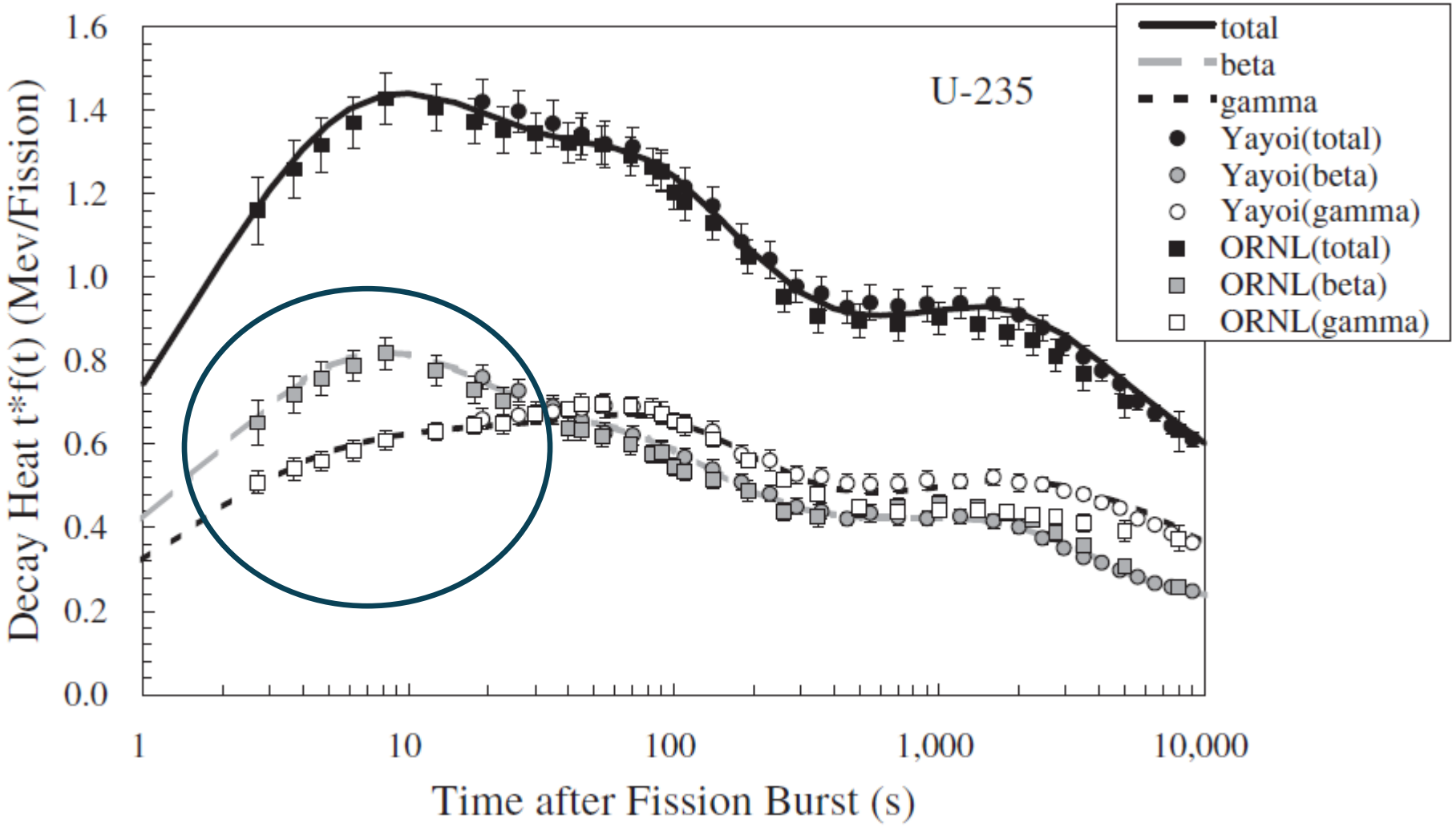


Electron detector

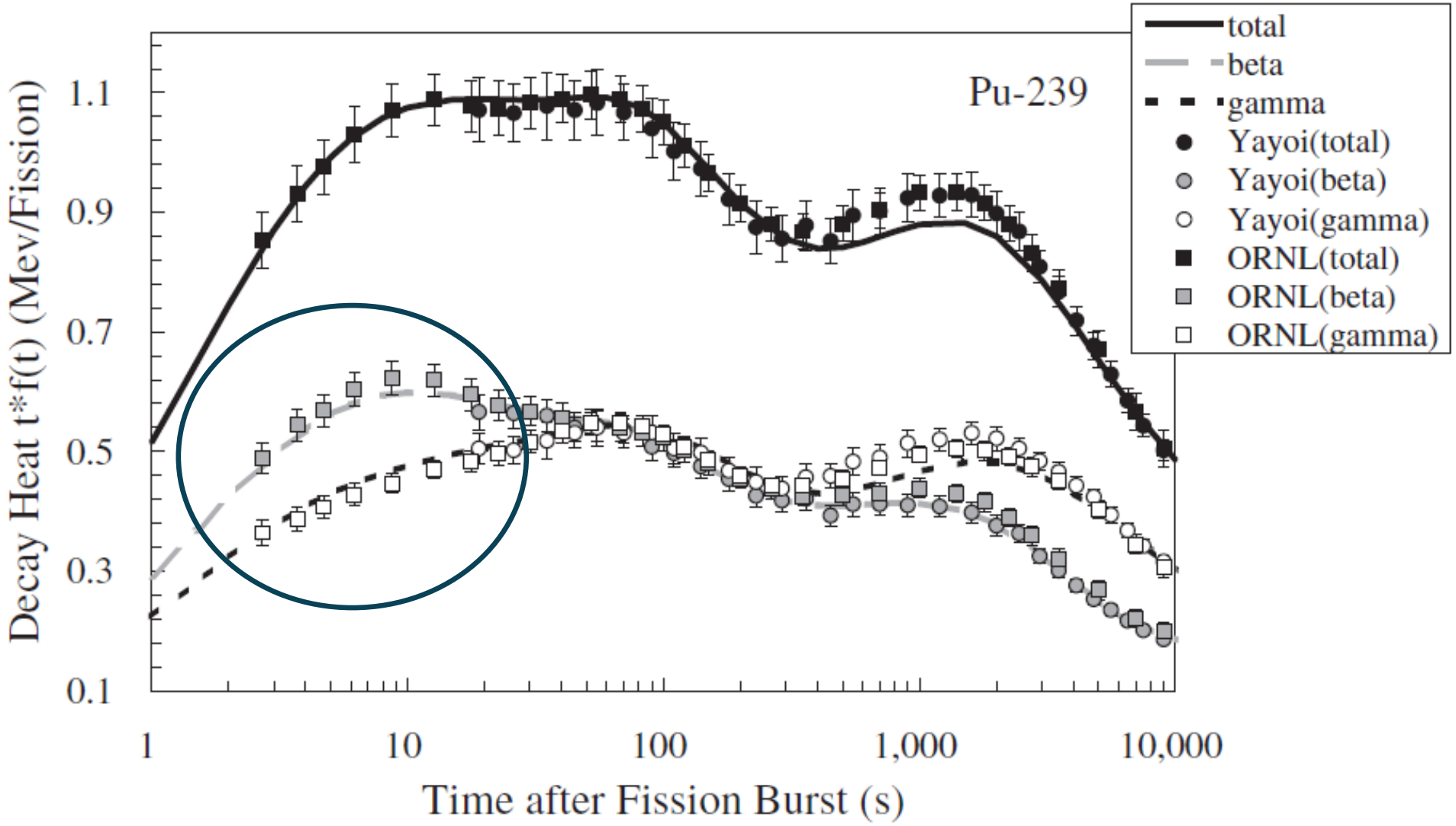


Brookhaven National Laboratory

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)

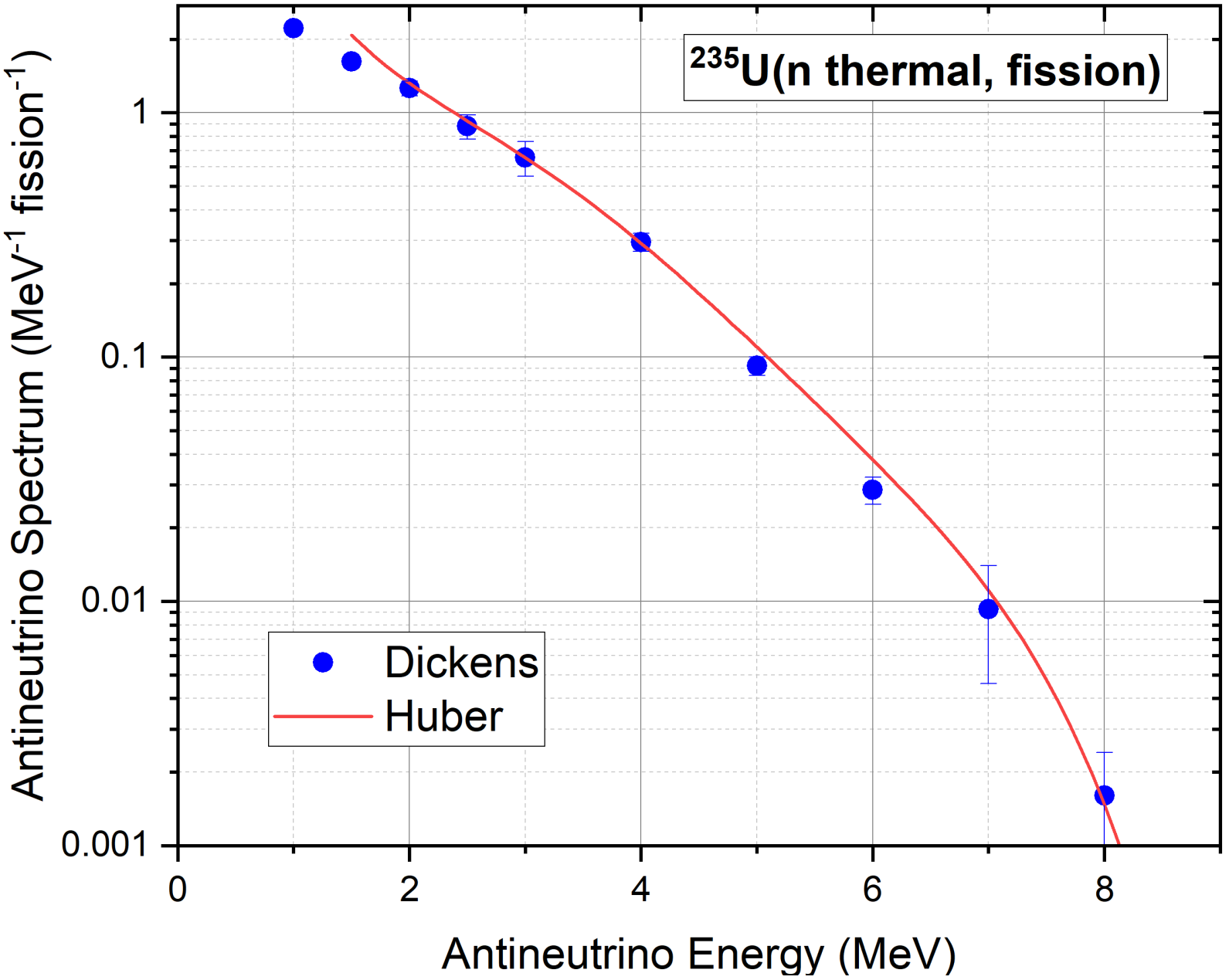


Reconsideration of the Theoretical Supplementation of Decay Data in Fission Product Decay Heat Summation Calculations
N. Hagura, T. Yoshida, T. Tachibana, Journal of Nuclear Science and Technology, 43:5, 497 (2012)

Brookhaven National Laboratory

Pioneer ^{235}U 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!



J. K. Dickens, Phys. Rev. Lett. 46, 1061 (1981).

Brookhaven National Laboratory

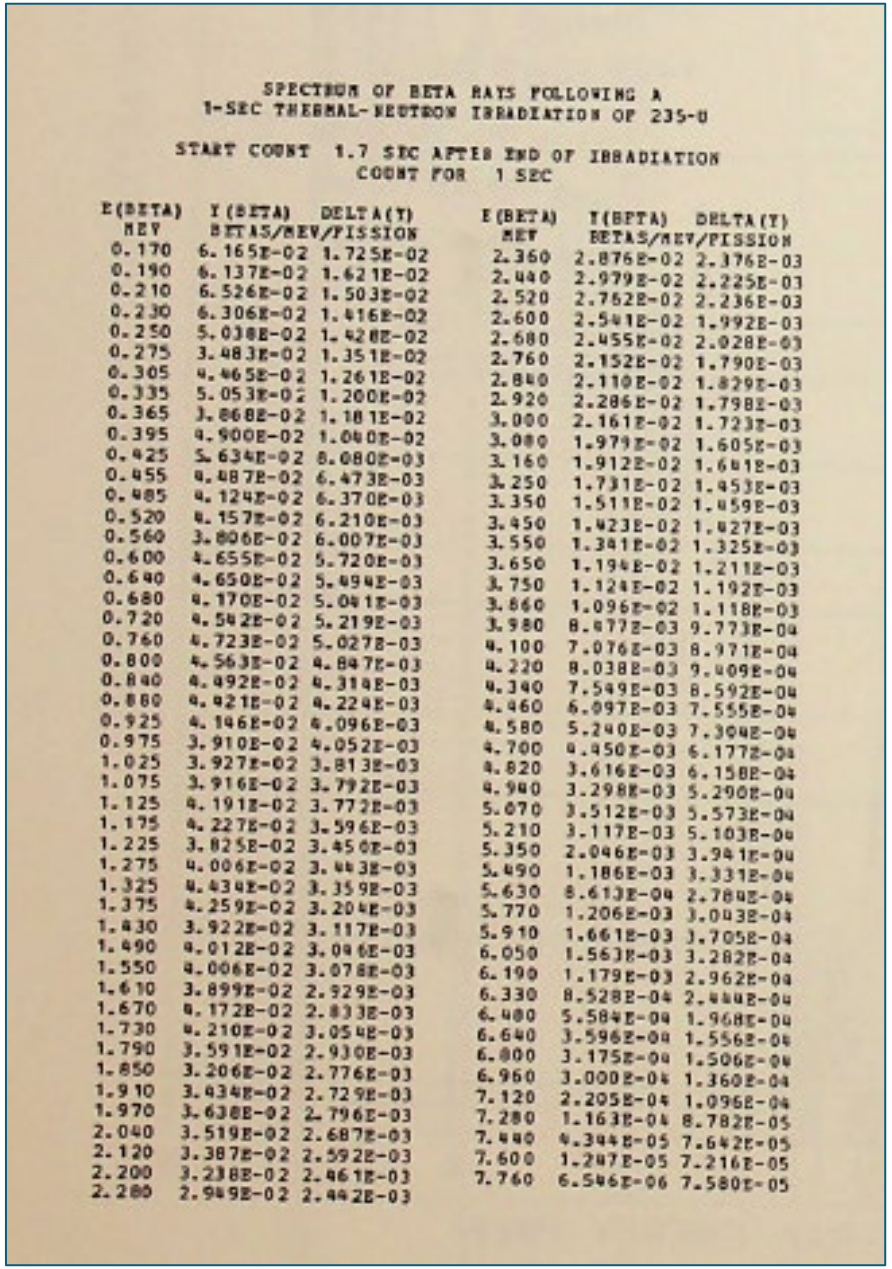
ORNL irradiations

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.

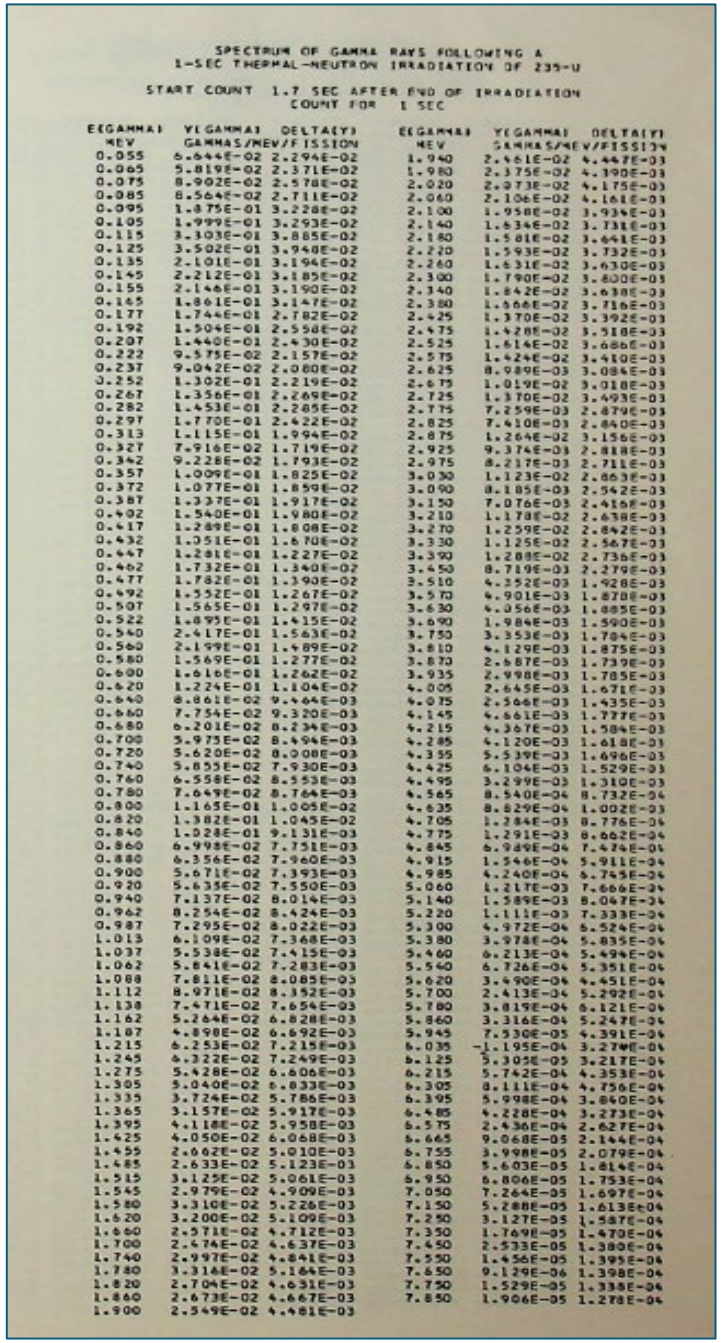
The three reports were also converted to PDF with a high-quality scanner.



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

START COUNT 1.7 SEC AFTER END OF IRRADIATION
COUNT FOR 1 SEC

E(BETA) MEV	I(BETA) BETAS/MEV/FISSION	DELTA(I)	E(BETA) MEV	I(BETA) BETAS/MEV/FISSION	DELTA(I)
0.170	6.165E-02	1.725E-02	2.360	2.876E-02	2.376E-03
0.190	6.137E-02	1.621E-02	2.440	2.979E-02	2.225E-03
0.210	6.526E-02	1.503E-02	2.520	2.762E-02	2.236E-03
0.230	6.306E-02	1.416E-02	2.600	2.541E-02	1.992E-03
0.250	5.038E-02	1.428E-02	2.680	2.455E-02	2.028E-03
0.275	3.483E-02	1.351E-02	2.760	2.152E-02	1.790E-03
0.305	4.465E-02	1.261E-02	2.840	2.110E-02	1.829E-03
0.335	5.053E-02	1.200E-02	2.920	2.286E-02	1.798E-03
0.365	3.868E-02	1.181E-02	3.000	2.161E-02	1.723E-03
0.395	4.900E-02	1.040E-02	3.080	1.979E-02	1.605E-03
0.425	5.634E-02	8.080E-03	3.160	1.912E-02	1.681E-03
0.455	4.487E-02	6.473E-03	3.250	1.731E-02	1.453E-03
0.485	4.124E-02	6.370E-03	3.350	1.511E-02	1.459E-03
0.520	4.157E-02	6.210E-03	3.450	1.423E-02	1.427E-03
0.560	3.806E-02	6.007E-03	3.550	1.341E-02	1.325E-03
0.600	4.655E-02	5.720E-03	3.650	1.194E-02	1.211E-03
0.640	4.650E-02	5.494E-03	3.750	1.124E-02	1.192E-03
0.680	4.170E-02	5.041E-03	3.860	1.096E-02	1.118E-03
0.720	4.542E-02	5.219E-03	3.980	8.877E-03	9.773E-04
0.760	4.723E-02	5.027E-03	4.100	7.076E-03	8.971E-04
0.800	4.563E-02	4.847E-03	4.220	8.038E-03	9.409E-04
0.840	4.492E-02	4.318E-03	4.340	7.549E-03	8.592E-04
0.880	4.421E-02	4.224E-03	4.460	6.097E-03	7.555E-04
0.925	4.146E-02	4.096E-03	4.580	5.240E-03	7.304E-04
0.975	3.910E-02	4.052E-03	4.700	4.450E-03	6.177E-04
1.025	3.927E-02	3.813E-03	4.820	3.616E-03	6.158E-04
1.075	3.916E-02	3.792E-03	4.940	3.298E-03	5.290E-04
1.125	4.191E-02	3.772E-03	5.070	3.512E-03	5.573E-04
1.175	4.227E-02	3.596E-03	5.210	3.117E-03	5.103E-04
1.225	3.825E-02	3.450E-03	5.350	2.046E-03	3.941E-04
1.275	4.006E-02	3.443E-03	5.490	1.186E-03	3.331E-04
1.325	4.434E-02	3.359E-03	5.630	8.613E-04	2.784E-04
1.375	4.259E-02	3.204E-03	5.770	1.206E-03	3.043E-04
1.430	3.922E-02	3.117E-03	5.910	1.661E-03	3.705E-04
1.490	4.012E-02	3.096E-03	6.050	1.563E-03	3.282E-04
1.550	4.006E-02	3.078E-03	6.190	1.179E-03	2.962E-04
1.610	3.899E-02	2.929E-03	6.330	8.528E-04	2.444E-04
1.670	4.172E-02	2.833E-03	6.480	5.584E-04	1.968E-04
1.730	4.210E-02	3.054E-03	6.640	3.596E-04	1.556E-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	3.000E-04	1.360E-04
1.910	3.434E-02	2.729E-03	7.120	2.205E-04	1.096E-04
1.970	3.638E-02	2.796E-03	7.280	1.163E-04	8.782E-05
2.040	3.519E-02	2.687E-03	7.440	4.344E-05	7.642E-05
2.120	3.387E-02	2.592E-03	7.600	1.247E-05	7.216E-05
2.200	3.238E-02	2.461E-03	7.760	6.546E-06	7.580E-05
2.280	2.949E-02	2.442E-03			



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

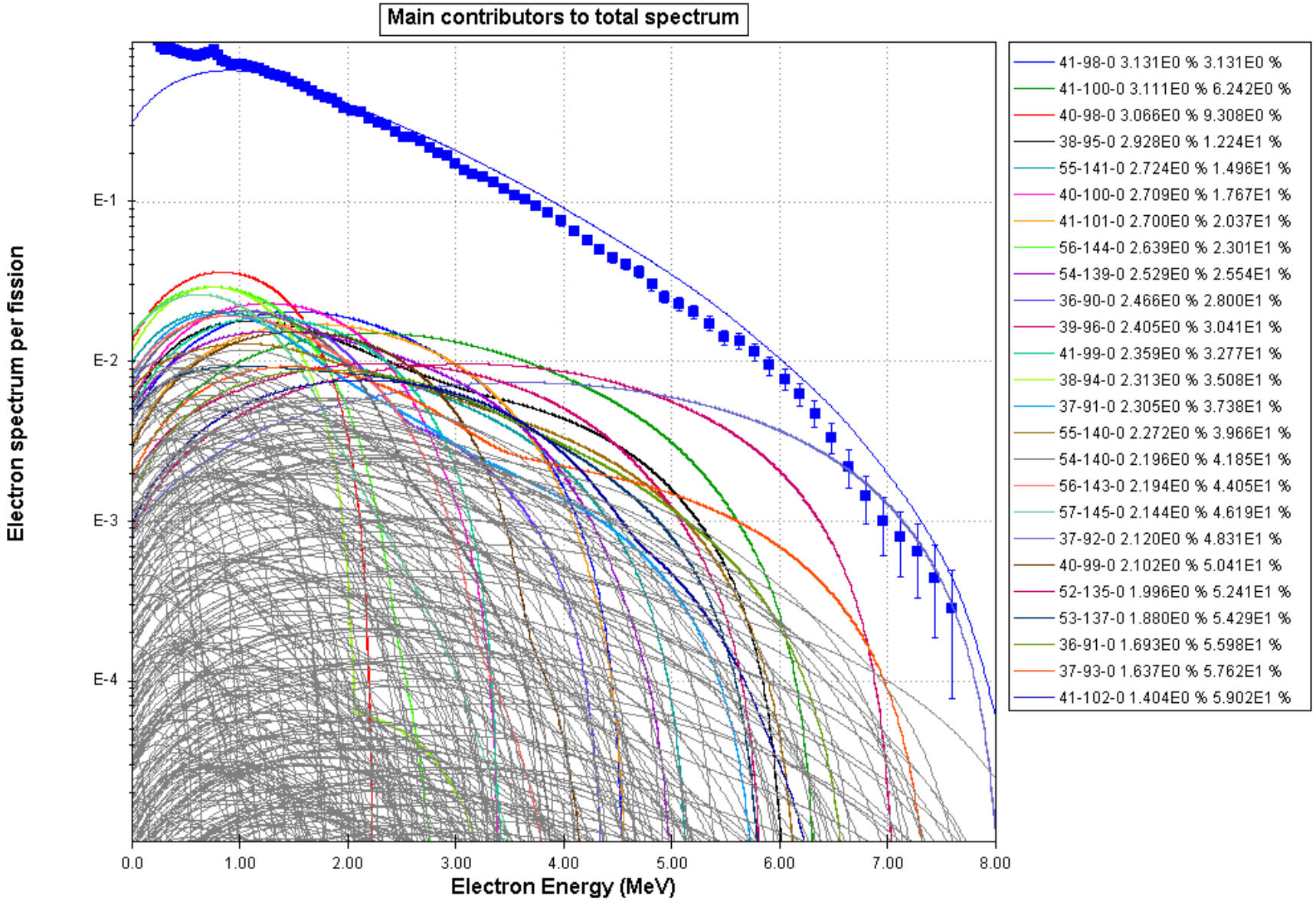
START COUNT 1.7 SEC AFTER END OF IRRADIATION
COUNT FOR 1 SEC

EGAMMA1 MEV	YGAMMA1 GAMMAS/MEV/FISSION	DELTA(Y)	EGAMMA1 MEV	YGAMMA1 GAMMAS/MEV/FISSION	DELTA(Y)
0.055	6.644E-02	2.294E-02	1.940	2.461E-02	4.447E-03
0.065	5.819E-02	2.371E-02	1.980	2.375E-02	4.390E-03
0.075	8.902E-02	2.578E-02	2.020	2.973E-02	4.175E-03
0.085	8.564E-02	2.711E-02	2.060	2.106E-02	4.161E-03
0.095	1.875E-01	3.228E-02	2.100	1.958E-02	3.934E-03
0.105	1.999E-01	3.293E-02	2.140	1.634E-02	3.731E-03
0.115	3.303E-01	3.885E-02	2.180	1.581E-02	3.641E-03
0.125	3.502E-01	3.948E-02	2.220	1.593E-02	3.732E-03
0.135	2.101E-01	3.194E-02	2.260	1.631E-02	3.630E-03
0.145	2.212E-01	3.185E-02	2.300	1.790E-02	3.800E-03
0.155	2.146E-01	3.190E-02	2.340	1.842E-02	3.638E-03
0.165	1.861E-01	3.147E-02	2.380	1.668E-02	3.716E-03
0.177	1.744E-01	2.782E-02	2.425	1.370E-02	3.392E-03
0.192	1.504E-01	2.558E-02	2.475	1.428E-02	3.518E-03
0.207	1.440E-01	2.430E-02	2.525	1.614E-02	3.686E-03
0.222	9.575E-02	2.157E-02	2.575	1.424E-02	3.410E-03
0.237	9.042E-02	2.080E-02	2.625	8.989E-03	3.084E-03
0.252	1.302E-01	2.219E-02	2.675	1.019E-02	3.018E-03
0.267	1.356E-01	2.269E-02	2.725	1.370E-02	3.493E-03
0.282	1.453E-01	2.285E-02	2.775	7.259E-03	2.879E-03
0.297	1.470E-01	2.422E-02	2.825	7.410E-03	2.840E-03
0.313	1.115E-01	1.994E-02	2.875	1.264E-02	3.156E-03
0.327	7.916E-02	1.719E-02	2.925	9.374E-03	2.818E-03
0.342	9.228E-02	1.793E-02	2.975	8.217E-03	2.711E-03
0.357	1.009E-01	1.825E-02	3.030	1.123E-02	2.863E-03
0.372	1.077E-01	1.859E-02	3.090	8.185E-03	2.542E-03
0.387	1.337E-01	1.917E-02	3.150	7.076E-03	2.415E-03
0.402	1.540E-01	1.980E-02	3.210	1.178E-02	2.638E-03
0.417	1.289E-01	1.808E-02	3.270	1.259E-02	2.842E-03
0.432	1.051E-01	1.670E-02	3.330	1.125E-02	2.567E-03
0.447	1.281E-01	1.727E-02	3.390	1.288E-02	2.736E-03
0.462	1.732E-01	1.340E-02	3.450	8.719E-03	2.279E-03
0.477	1.782E-01	1.390E-02	3.510	4.352E-03	1.928E-03
0.492	1.552E-01	1.267E-02	3.570	4.901E-03	1.870E-03
0.507	1.565E-01	1.297E-02	3.630	4.056E-03	1.885E-03
0.522	1.895E-01	1.415E-02	3.690	1.984E-03	1.590E-03
0.540	2.417E-01	1.563E-02	3.750	3.353E-03	1.784E-03
0.560	2.199E-01	1.489E-02	3.810	4.129E-03	1.875E-03
0.580	1.569E-01	1.277E-02	3.870	2.687E-03	1.739E-03
0.600	1.616E-01	1.262E-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	8.861E-02	9.944E-03	4.075	2.566E-03	1.435E-03
0.660	7.754E-02	9.320E-03	4.145	4.661E-03	1.777E-03
0.680	6.201E-02	8.234E-03	4.215	4.367E-03	1.584E-03
0.700	5.975E-02	8.494E-03	4.285	4.120E-03	1.618E-03
0.720	5.620E-02	8.008E-03	4.355	5.539E-03	1.696E-03
0.740	5.855E-02	7.930E-03	4.425	6.104E-03	1.529E-03
0.760	6.558E-02	8.553E-03	4.495	3.299E-03	1.310E-03
0.780	7.649E-02	8.764E-03	4.565	8.540E-04	8.732E-04
0.800	1.165E-01	1.005E-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	1.028E-01	9.131E-03	4.775	1.291E-03	8.662E-04
0.860	6.998E-02	7.751E-03	4.845	6.989E-04	7.474E-04
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-04
0.940	7.137E-02	8.014E-03	5.140	1.589E-03	8.047E-04
0.962	8.254E-02	8.424E-03	5.220	1.111E-03	7.333E-04
0.987	7.295E-02	8.022E-03	5.300	4.972E-04	6.524E-04
1.013	6.109E-02	7.368E-03	5.380	3.978E-04	5.835E-04
1.037	5.538E-02	7.415E-03	5.460	6.213E-04	5.496E-04
1.062	5.841E-02	7.283E-03	5.540	6.726E-04	5.351E-04
1.088	7.811E-02	8.085E-03	5.620	3.490E-04	4.451E-04
1.112	8.971E-02	8.352E-03	5.700	2.413E-04	5.292E-04
1.138	7.471E-02	7.654E-03	5.780	3.819E-04	6.121E-04
1.162	5.264E-02	6.828E-03	5.860	3.316E-04	5.247E-04
1.187	4.898E-02	6.692E-03	5.945	7.530E-05	4.391E-04
1.215	6.253E-02	7.215E-03	6.035	-1.195E-04	3.279E-04
1.245	6.322E-02	7.249E-03	6.125	5.305E-05	3.217E-04
1.275	5.428E-02	6.606E-03	6.215	5.742E-04	4.353E-04
1.305	5.040E-02	6.833E-03	6.305	8.111E-04	4.756E-04
1.335	3.724E-02	5.786E-03	6.395	5.998E-04	3.840E-04
1.365	3.157E-02	5.917E-03	6.485	4.228E-04	3.273E-04
1.395	4.118E-02	5.958E-03	6.575	2.436E-04	2.627E-04
1.425	4.050E-02	6.068E-03	6.665	9.068E-05	2.144E-04
1.455	2.662E-02	5.010E-03	6.755	3.998E-05	2.079E-04
1.485	2.633E-02	5.123E-03	6.850	5.603E-05	1.814E-04
1.515	3.125E-02	5.061E-03	6.950	6.806E-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	1.613E-04
1.620	3.200E-02	5.109E-03	7.250	3.127E-05	1.587E-04
1.660	2.571E-02	4.712E-03	7.350	1.769E-05	1.470E-04
1.700	2.474E-02	4.637E-03	7.450	2.533E-05	1.390E-04
1.740	2.997E-02	4.841E-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	1.529E-05	1.338E-04
1.860	2.673E-02	4.667E-03	7.850	1.906E-05	1.278E-04
1.900	2.549E-02	4.481E-03			

Brookhaven National Laboratory

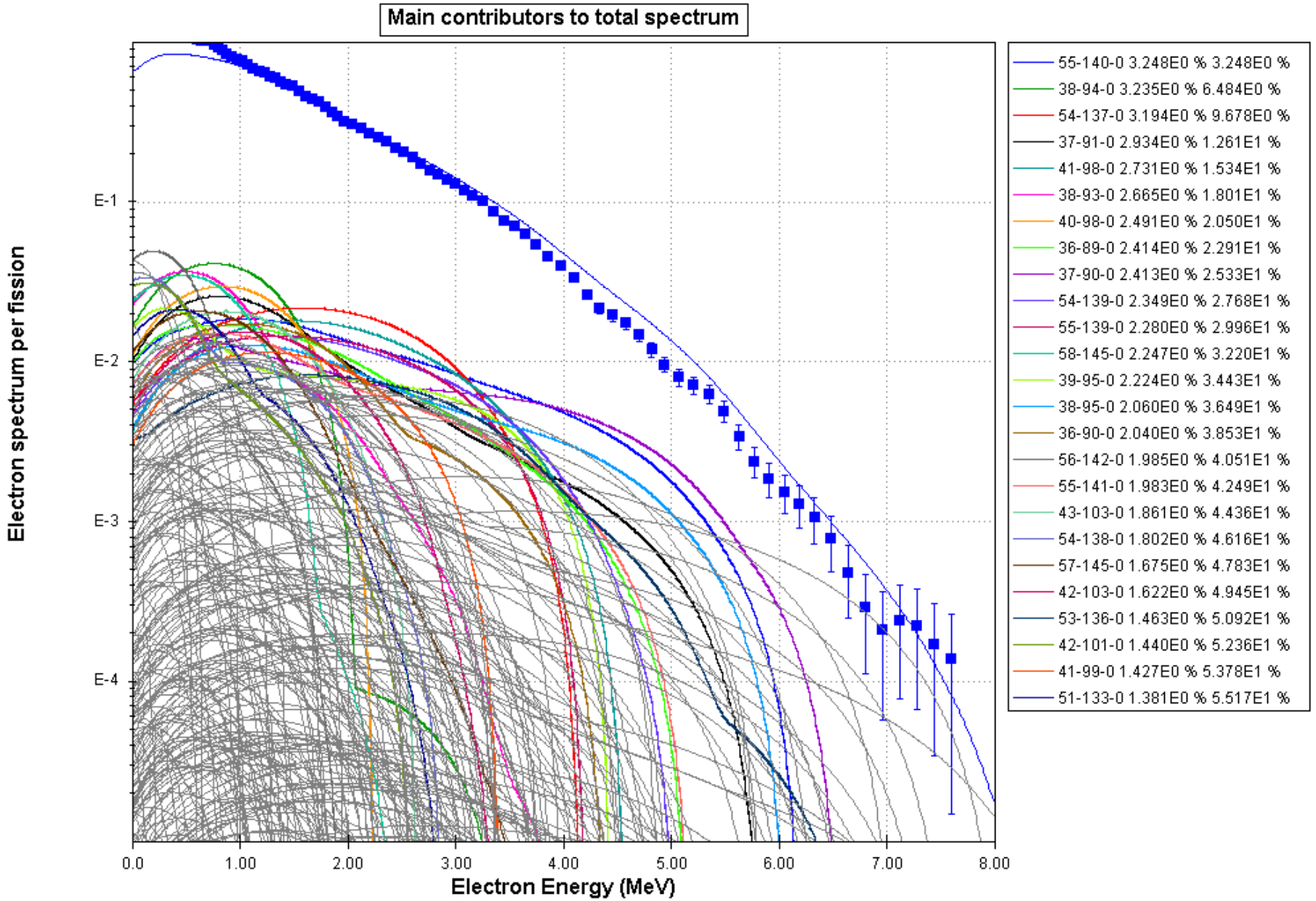
Delayed Electrons

Delayed electron data – short ^{235}U irradiation



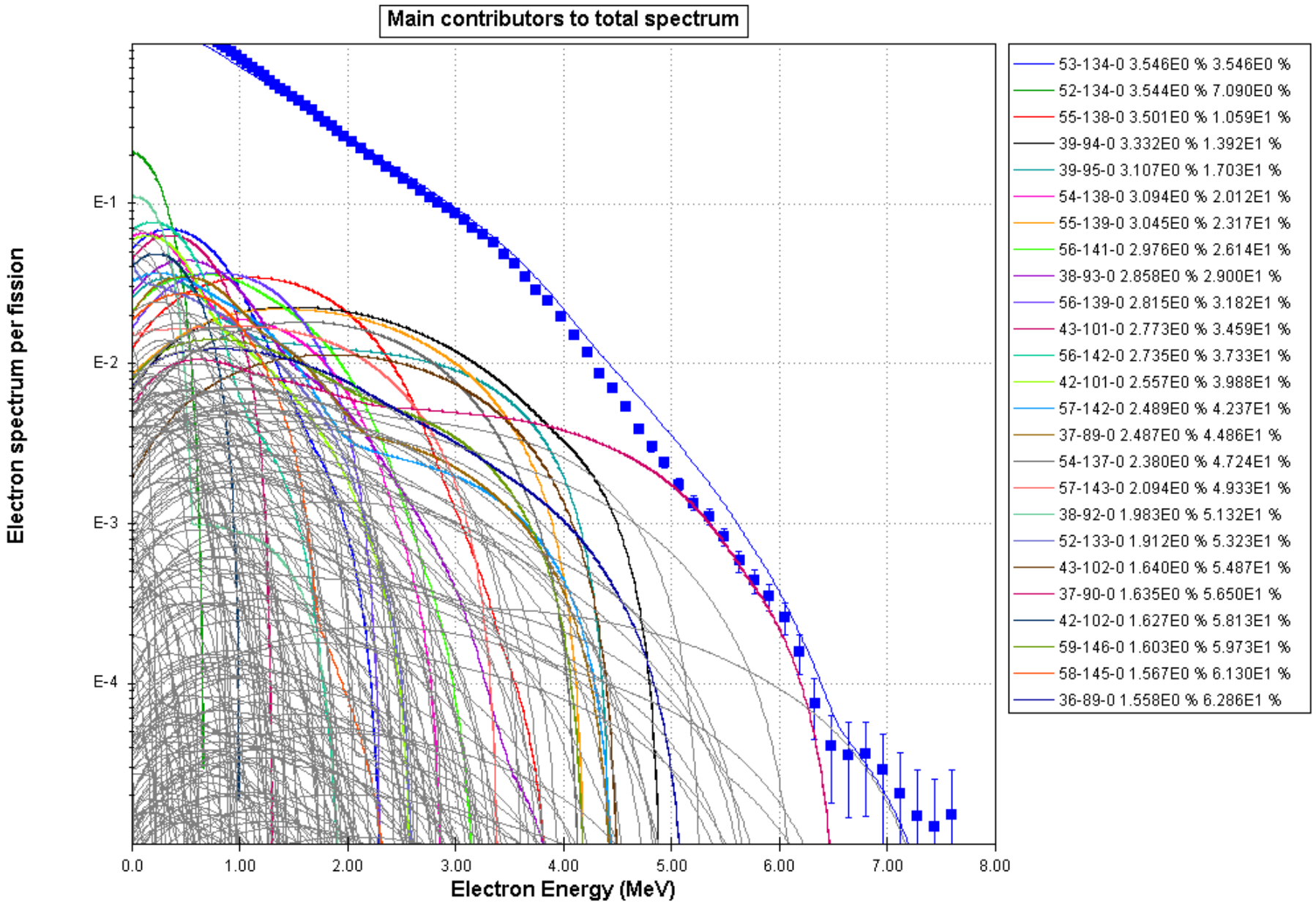
- We can clearly see that the ^{92}Rb and ^{96}Y contributions are clearly too large.
- Since their decay data are well known, we can adjust their independent fission yield to match the data.

Delayed electron data – medium ^{235}U irradiation



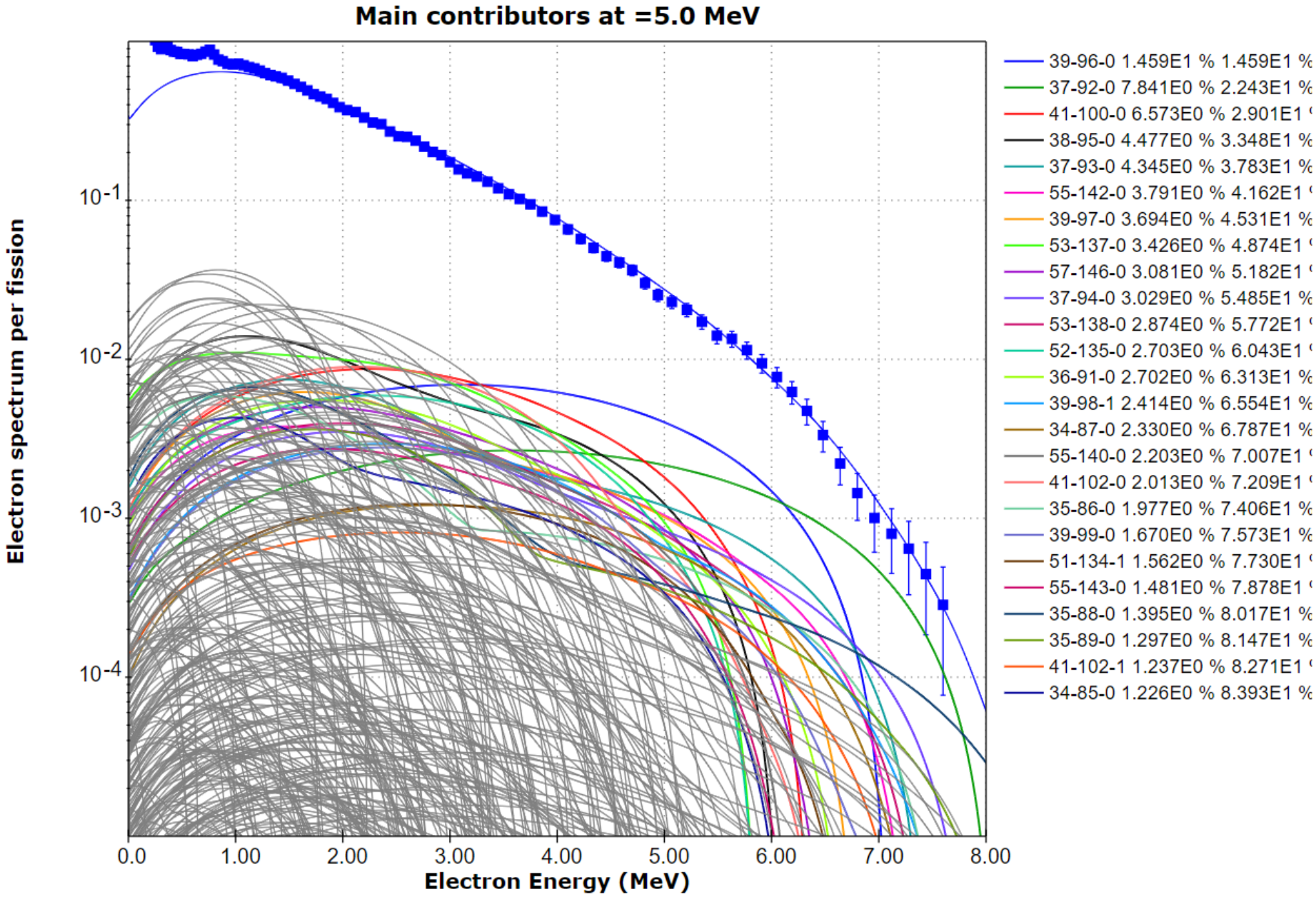
- Similarly, for ^{140}Cs and ^{90}Rb.

Delayed electron data – long ^{235}U irradiation



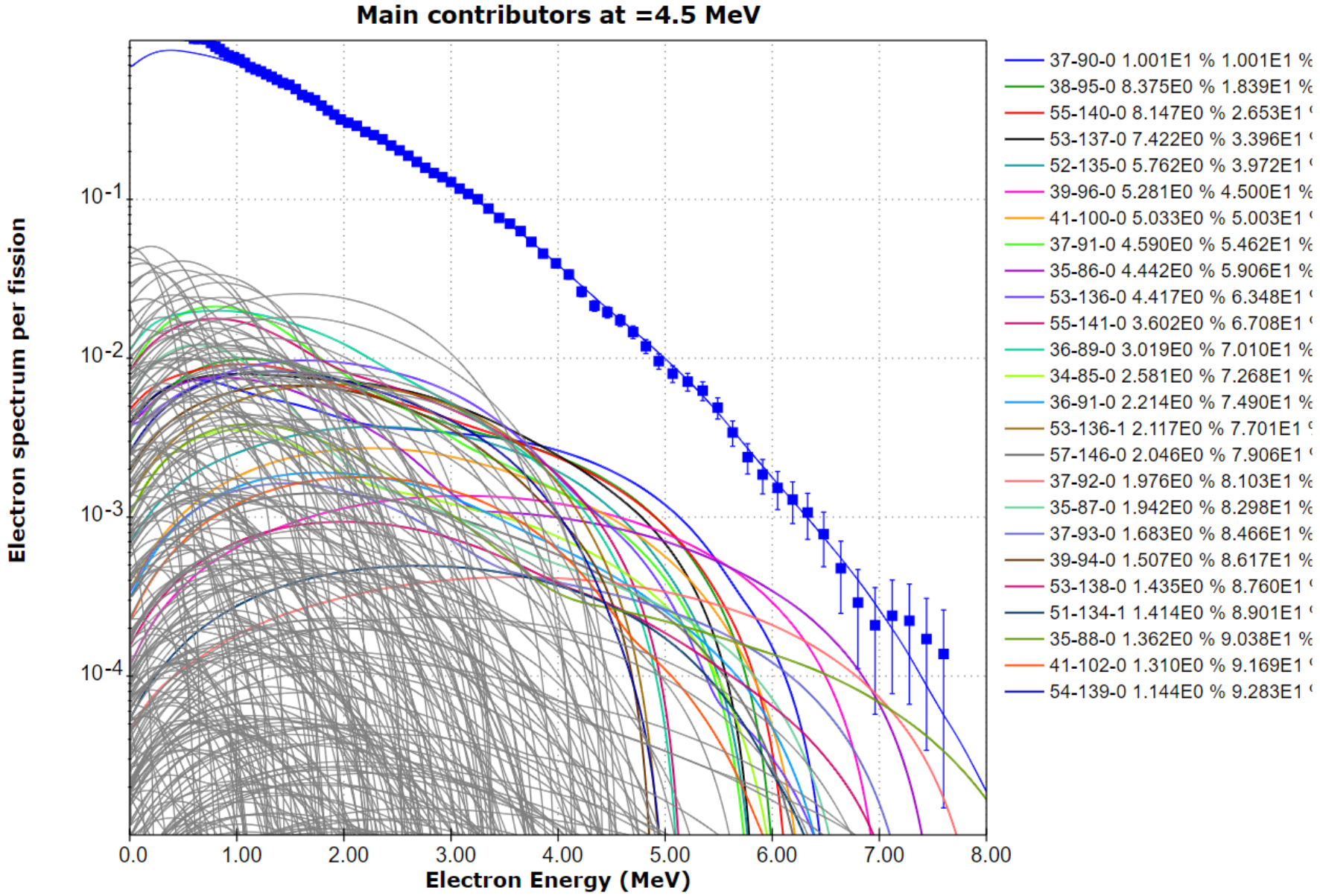
- Similarly, for ^{101}Tc and ^{94}Y.

Short ^{235}U irradiation – adjusted yields



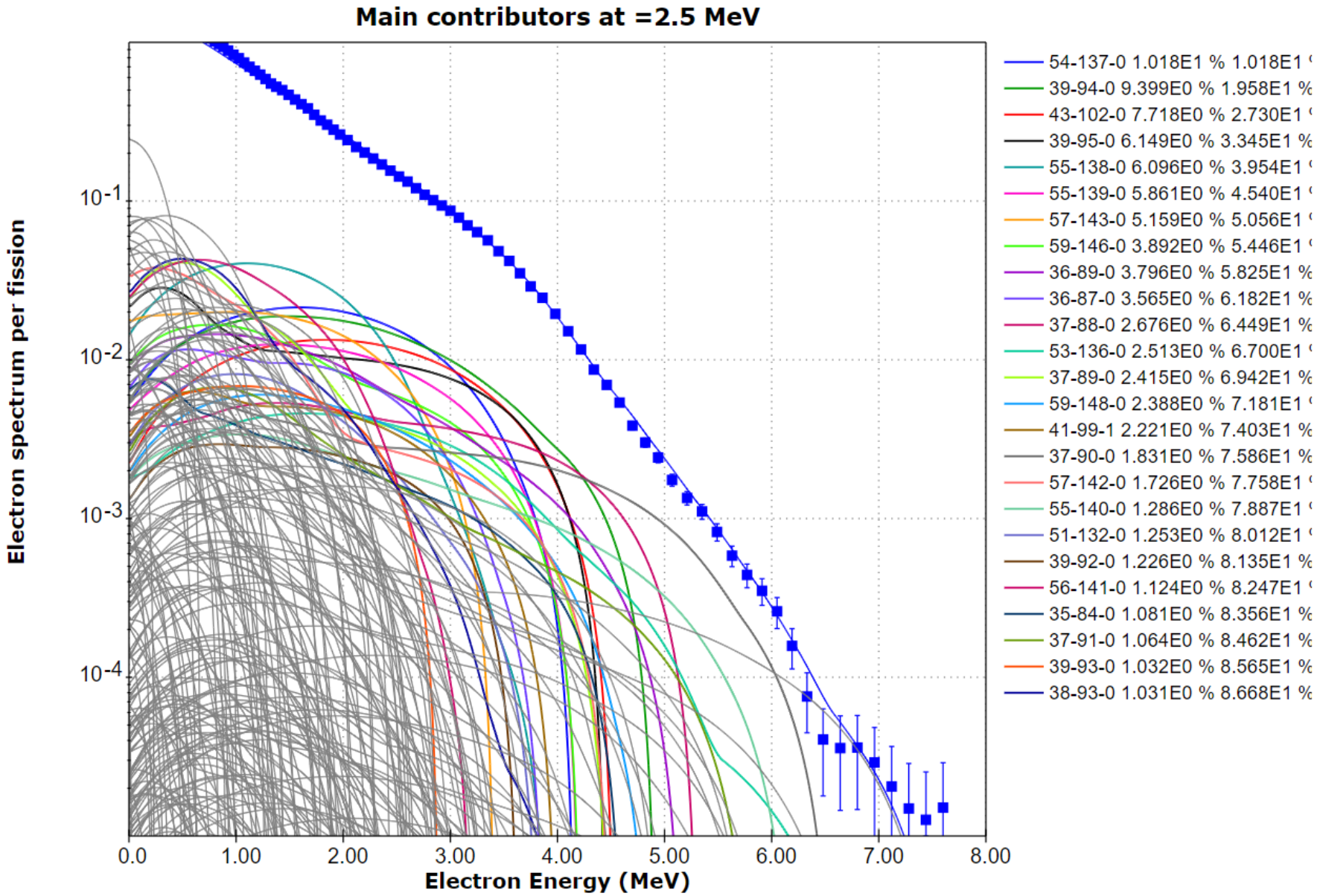
Brookhaven National Laboratory

Medium ^{235}U irradiation – adjusted yields

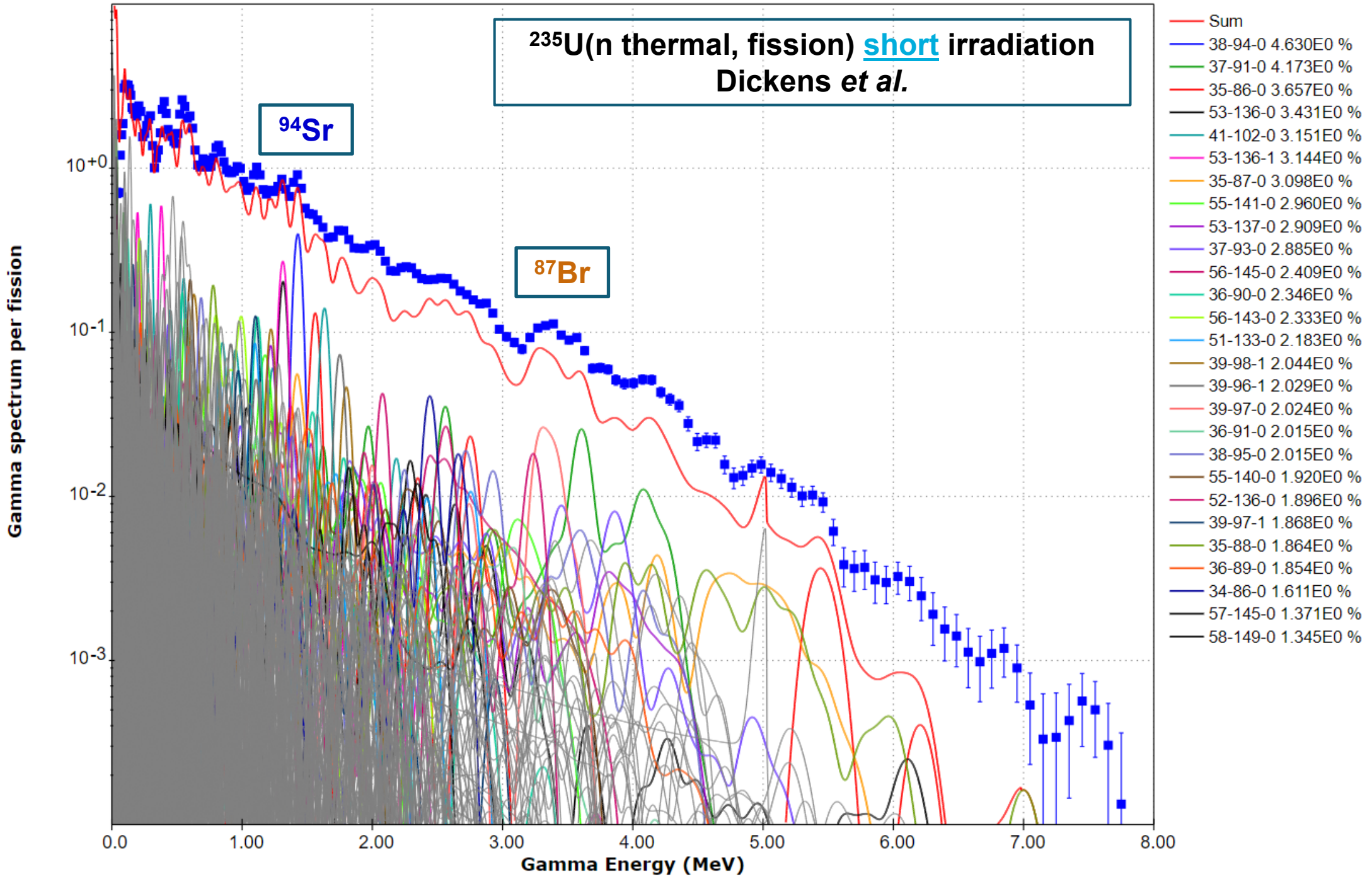


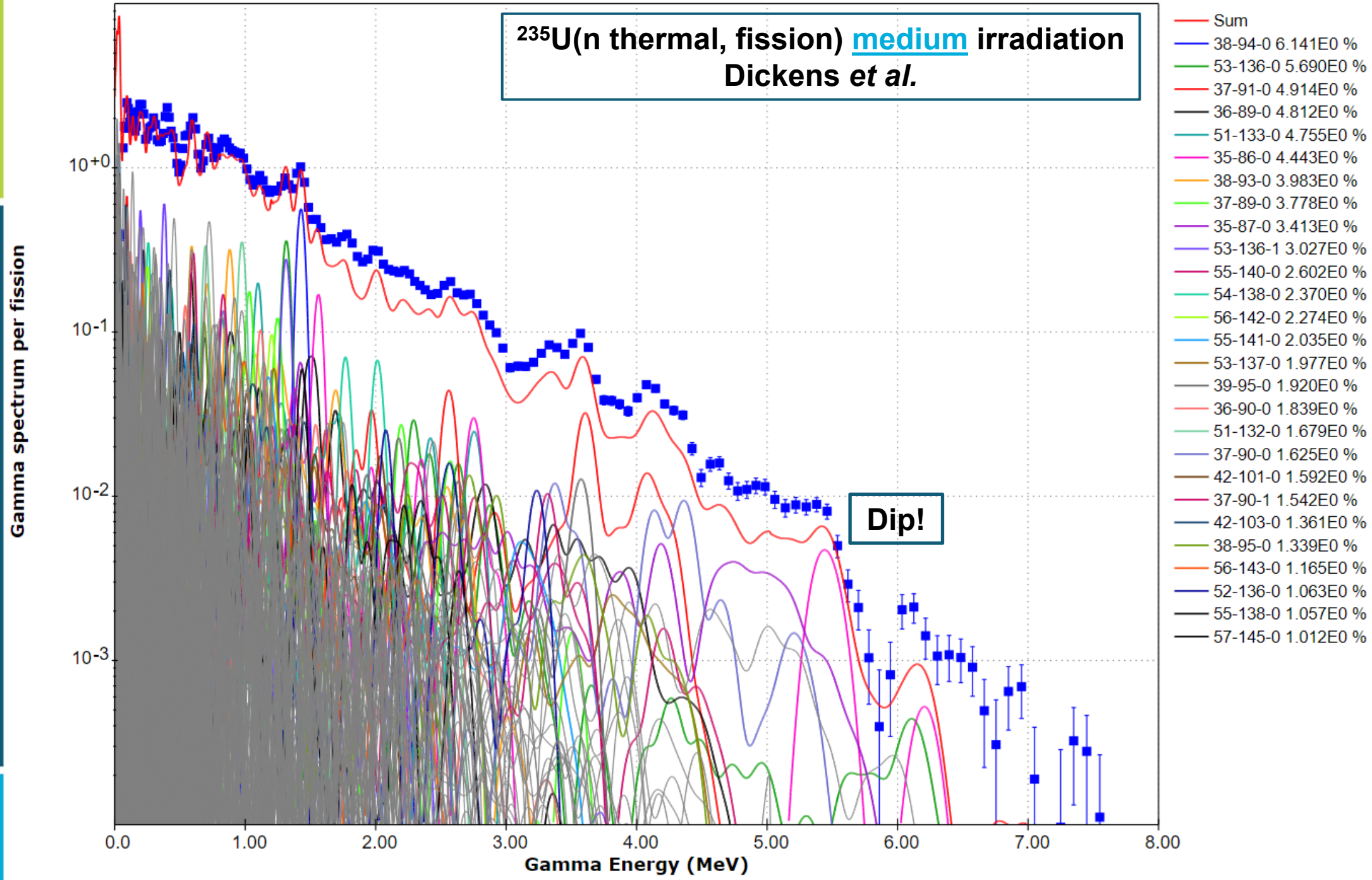
Brookhaven National Laboratory

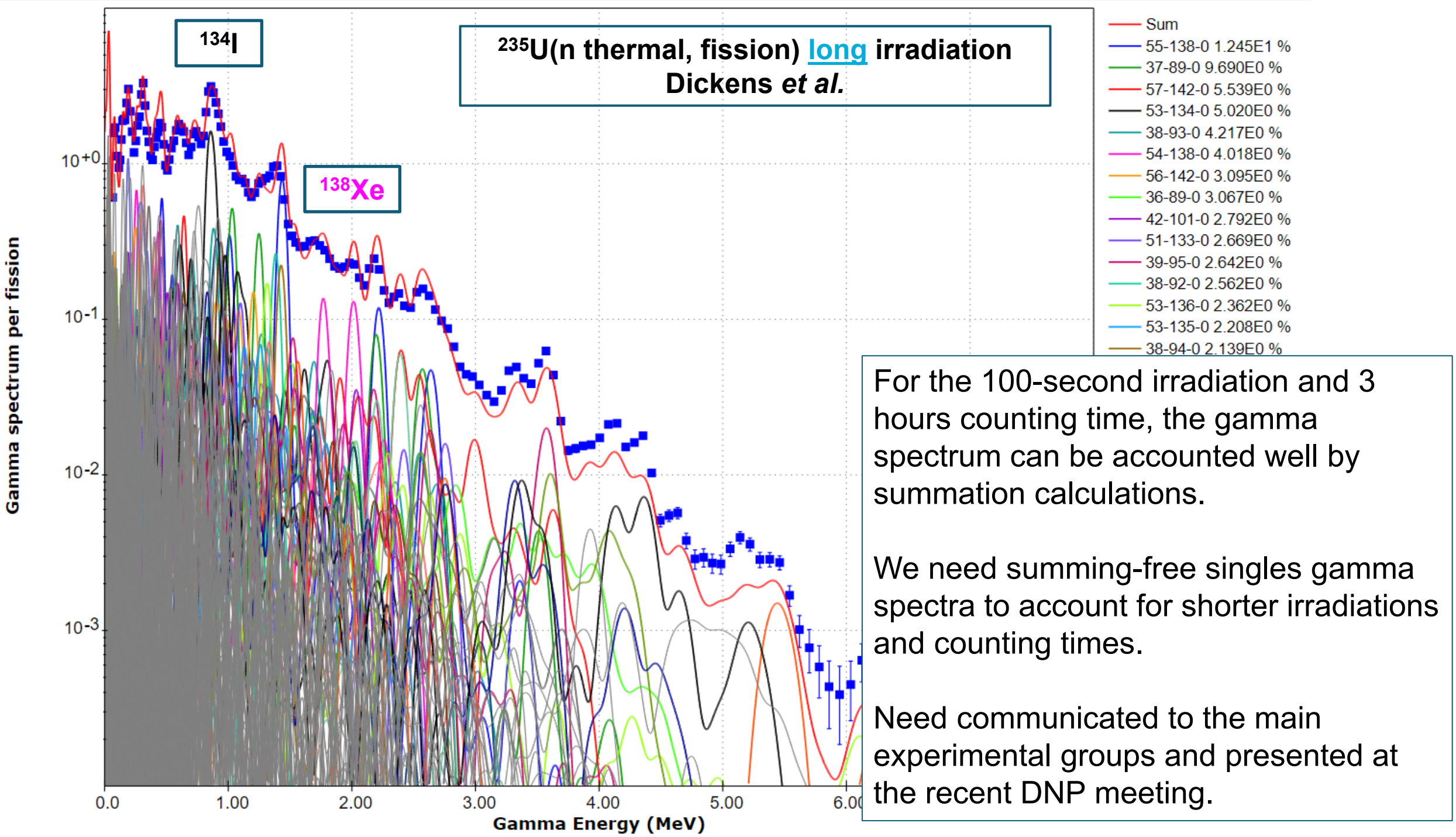
Long ^{235}U irradiation – adjusted yields



Brookhaven National Laboratory


235U(n thermal, fission) short irradiation
Dickens et al.
94Sr
87Br
Gamma spectrum per fission
10+0
10-1
10-2
10-3
0.0
1.00
2.00
3.00
4.00
5.00
6.00
7.00
8.00
Gamma Energy (MeV)
Sum
38-94-0 4.630E0 %
37-91-0 4.173E0 %
35-86-0 3.657E0 %
53-136-0 3.431E0 %
41-102-0 3.151E0 %
53-136-1 3.144E0 %
35-87-0 3.098E0 %
55-141-0 2.960E0 %
53-137-0 2.909E0 %
37-93-0 2.885E0 %
56-145-0 2.409E0 %
36-90-0 2.346E0 %
56-143-0 2.333E0 %
51-133-0 2.183E0 %
39-98-1 2.044E0 %
39-96-1 2.029E0 %
39-97-0 2.024E0 %
36-91-0 2.015E0 %
38-95-0 2.015E0 %
55-140-0 1.920E0 %
52-136-0 1.896E0 %
39-97-1 1.868E0 %
35-88-0 1.864E0 %
36-89-0 1.854E0 %
34-86-0 1.611E0 %
57-145-0 1.371E0 %
58-149-0 1.345E0 %


235U(n thermal, fission) medium irradiation
Dickens et al.
Dip!
Gamma spectrum per fission
Gamma Energy (MeV)
10+0
10-1
10-2
10-3
0.0
1.00
2.00
3.00
4.00
5.00
6.00
7.00
8.00
Sum
38-94-0 6.141E0 %
53-136-0 5.690E0 %
37-91-0 4.914E0 %
36-89-0 4.812E0 %
51-133-0 4.755E0 %
35-86-0 4.443E0 %
38-93-0 3.983E0 %
37-89-0 3.778E0 %
35-87-0 3.413E0 %
53-136-1 3.027E0 %
55-140-0 2.602E0 %
54-138-0 2.370E0 %
56-142-0 2.274E0 %
55-141-0 2.035E0 %
53-137-0 1.977E0 %
39-95-0 1.920E0 %
36-90-0 1.839E0 %
51-132-0 1.679E0 %
37-90-0 1.625E0 %
42-101-0 1.592E0 %
37-90-1 1.542E0 %
42-103-0 1.361E0 %
38-95-0 1.339E0 %
56-143-0 1.165E0 %
52-136-0 1.063E0 %
55-138-0 1.057E0 %
57-145-0 1.012E0 %



For the 100-second irradiation and 3 hours counting time, the gamma spectrum can be accounted well by summation calculations.

We need summing-free singles gamma spectra to account for shorter irradiations and counting times.

Need communicated to the main experimental groups and presented at the recent DNP meeting.

Conclusions

- We are trying to understand the reduction in Independent Fission Yields for about 10 isotopes, which consistently appear in the three fissioning systems. Working with Andrea Mattera to check if there are measurements for these yields or are based on **P(Z/A) x Y(A).**

- Adjustment to delayed electron activity is clearly a powerful tool to obtain independent fission yields for short-lived fission products, not surprisingly since highly precise delayed neutron activities have been used with that purpose in the past.

- Our capability to predict delayed gamma spectra at short times after fission, less than several hundred seconds, can be improved.

- We need gamma spectra for short-lived fission products using high-efficiency detector setups. Communicated this need with the leading TAGS experimental groups. Perhaps data from segmented detectors could be re-analyzed with this purpose.

- A couple of reports will be written soon detailing this work.

Grateful to the Zharia Harris, Becket Hill, Bryan Palaguachi and Matthew Seeley, as well to Catherine Dunn, who contributed greatly to this project!