LLNL-PRES-460638

Lawrence Livermore National Laboratory

Delayed Fission Gammas

Ed Lent and Dave Heinrichs

Presented at the Cross Section Evaluation Working Group (CSEWG) Meeting
in Santa Fe, NM, November 1-4, 2010

Lawrence Livermore National Laboratory, P.O. Box 808, L-198, Livermore, CA 94551-0808

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344

Introduction

LLNL has a DOE NCSP task to test the suitability of ENDF/B-VII.0 delayed fission gammas data for use in criticality accident (dose) assessment

* ENDF/B-VII.0 delayed fission gamma data are not suitable for this application (slides 3 and 4)

* Yanagisawa's results – Journal of Nuclear Science and Technology, vol. 39, no. 5, 499-505 (2002) – look promising

* **New work by Ed Lent (LLNL)** following Pruett and Yanagisawa

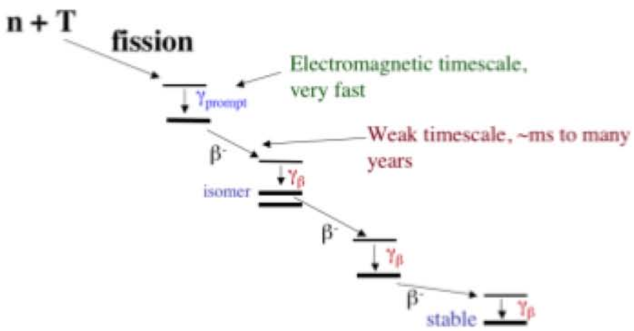
* **Proposed new data for ENDF/B-VII.1**

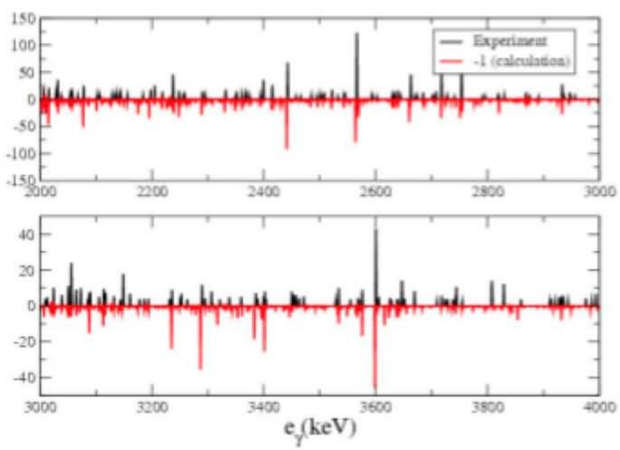
ENDF/B-VII.0

ENDF/B-VII.0 data provided by LLNL

UNCLASSIFIED

Simulating β-delayed γ's from fission





Monte-Carlo model (J.Pruet, *et al.* NIM A, 521, 608 (2004))

- Generate fission fragment from England and Rider.
- Follow β-decay chain to stability
- Collect γ's along the way.
- Generally good agreement w/ experiment of Norman et al.

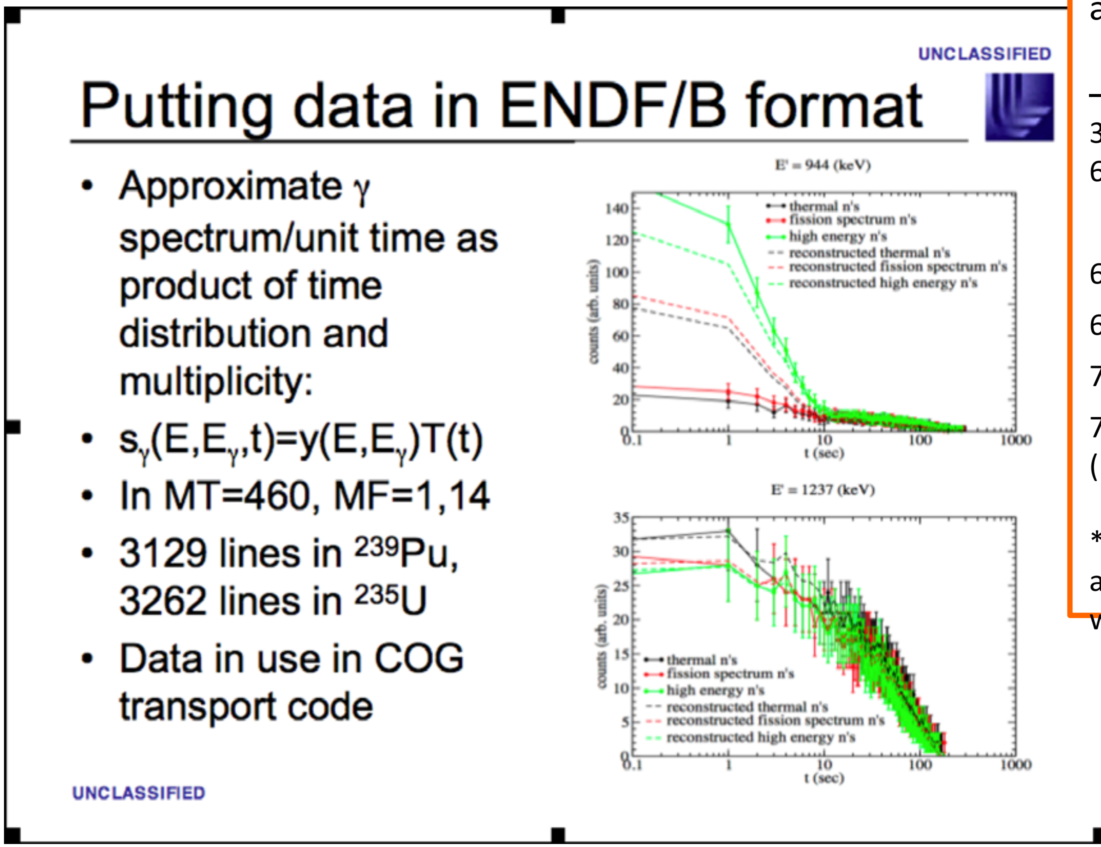
Monte Carlo

NuDat database

This detailed data should work for dose assessment purposes as well ….. but

UCRL-PRES-225819

Pruet 2004



Unfortunately, Pruet data way off in energy and multiplicity*:

Nd	Ed	Reference
3.6 g/f	2.89 MeV/f	Pruet (2004)
6.66 g/f	6.22 MeV/f	Lent (2010) ... this work
N/A	6.33(5) MeV/f	ENDF/B-VII.0
6.51 g/f	6.43(30) MeV/f	PhysRevC 6:1023(1972)
6.7 g/f	6.51(30) MeV/f	PhysRevC 7:1173 (1973)
7.45 g/f	7.18(26) MeV/f	PhysRevC 3:373 (1971)
7.9 g/f (1967)		PhysRevC 6:1023

*Personal communication with Dave Brown (N-Div) at mini-CSEWG (2010). Ed/Nd = 0.80 MeV/g (Pruet) when it should be ~ 1 MeV/g.

ENDF/B-VII.0 data is not suitable for dose calculations

Yanagisawa 2002

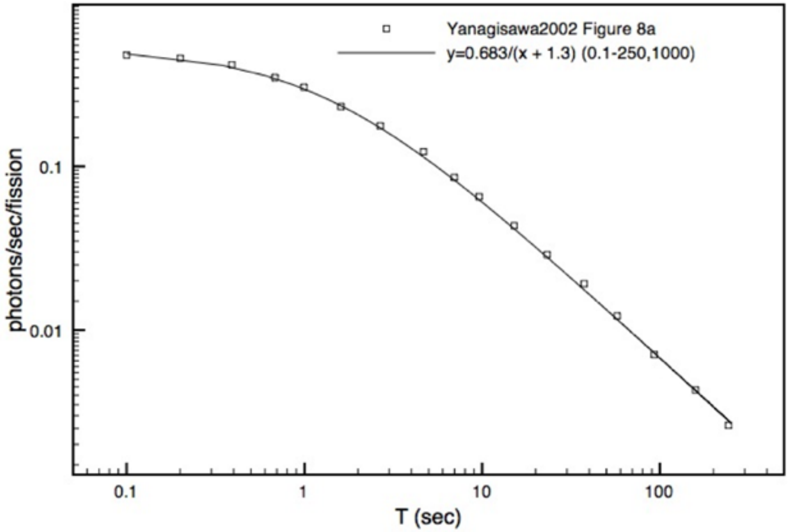
Journal of NUCLEAR SCIENCE and TECHNOLOGY, Vol. 39, No. 5, 499–505 (May 2002) provides detailed multiplicity data without spectra.

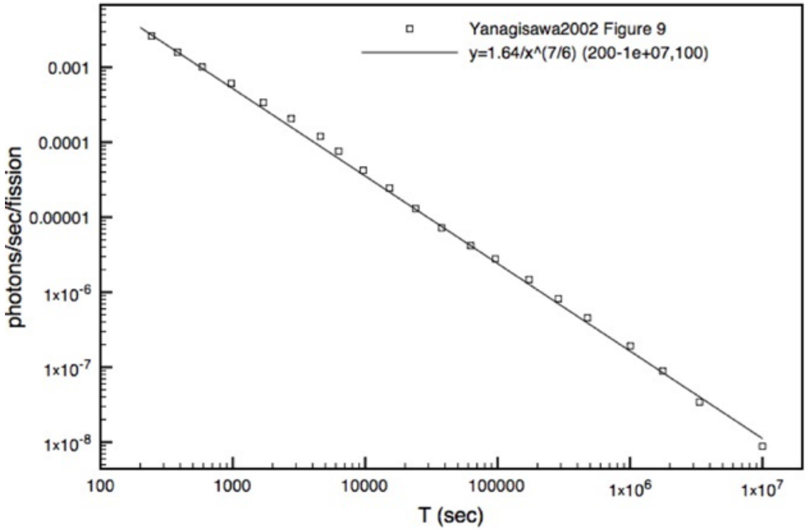
Ed Lent digitized and fit Yanagisawa's time-dependent photon multiplicity data

Way-Wigner $T^{-1.2}$

$n_d = 0.683/(T + 1.3)$, $T < 200$ sec

$n_d = 1.64/T^{7/6}$, $T > 200$ sec





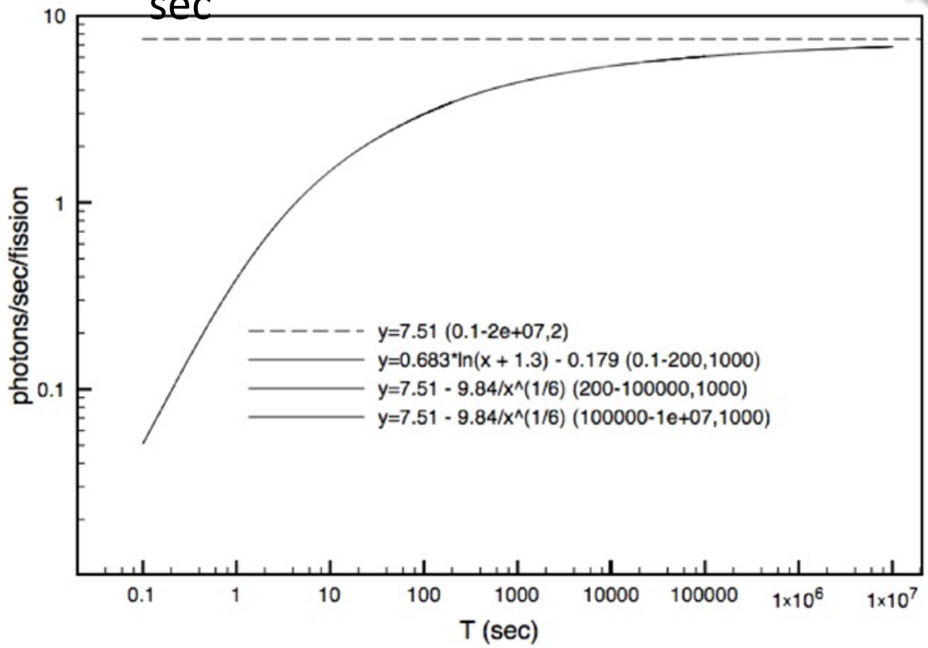
Yanagisawa 2002

Journal of NUCLEAR SCIENCE and TECHNOLOGY, Vol. 39, No. 5, 499–505 (May 2002)

Ed Lent also integrated these equations to determine the cumulative number of delayed photons in the time interval [0, T]:

$N_d = 0.683 \ln(T + 1.3) – 0.179$, T < 200 sec

$N_d = 7.51 – 9.84/T^{1/6}$, T > 200 sec



7.51 delayed photons per fission [0, ∞], which is in agreement with the value 7.45 ± 0.35 given by Peele and Maienschein, Phys. Rev. C 3:373 (1971).

This work

Ed Lent replicated previous work by Pruett and Yanagisawa by:

COGFY

T.R. England and B.F. Rider database (LA-UR-94-3106, ENDF-349) of fission products (789 FP nuclides for ^{235}U vs. probability at the instant of fission) **– like Pruett**

COGDC

JENDL-FPDD2000 containing 1221 FP half-lives, daughter branching ratios, and discrete and/or continuous gamma energy spectra **– like Yanagisawa**

RadSrc

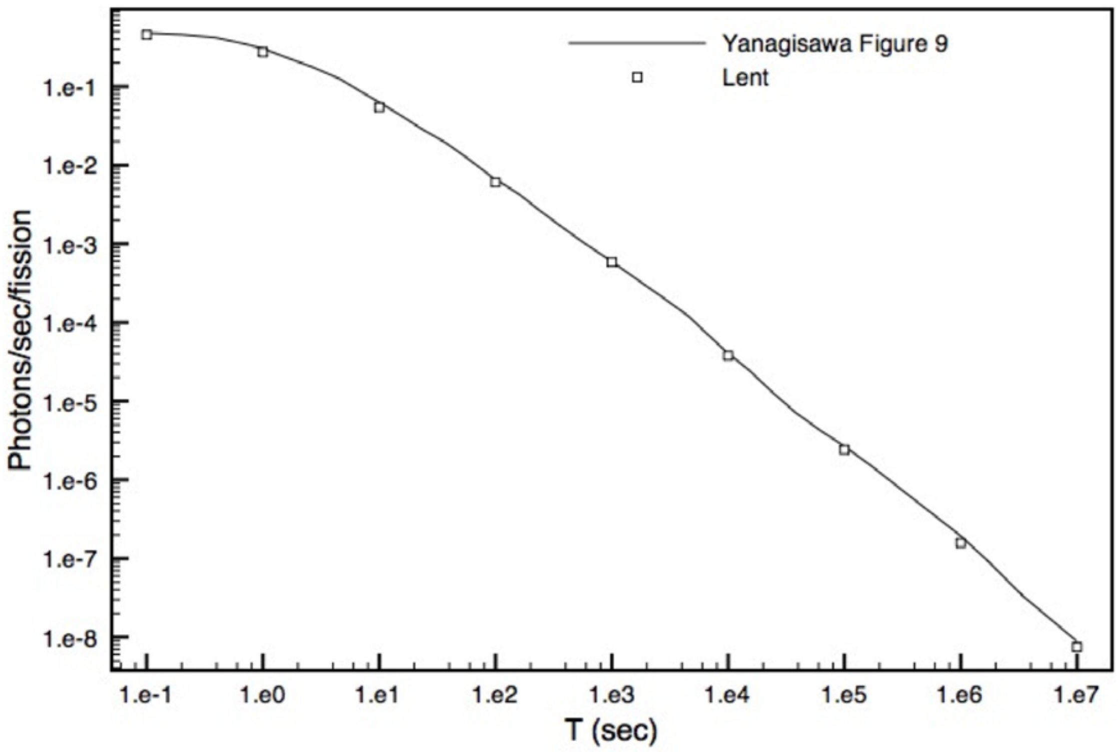
* Bateman solutions previously developed for gamma emission from α-decay

GetEG

The final piece is a code, GetEG, to put all the above pieces together. The input is ***isotope t***, where ***isotope*** is one of the FY isotopes (e.g., fast pooled neutron induced fission of U235), and ***t*** is the time in seconds. Start with the first nuclide in the ***isotope***'s fission product list. Use the DC branching ratio data to develop the (various possible) decay chain(s), add the DC half-life data to get the resultant nuclides amplitudes and decay rates at time ***t***, sum the DC discrete and/or continuous gamma energies to get the number of delayed gammas and associated gamma energy spectrum. Weight these results with the appropriate FY probability. Step through the remaining nuclides in the ***isotope***'s fission product list in a similar manner, summing the results as you go.

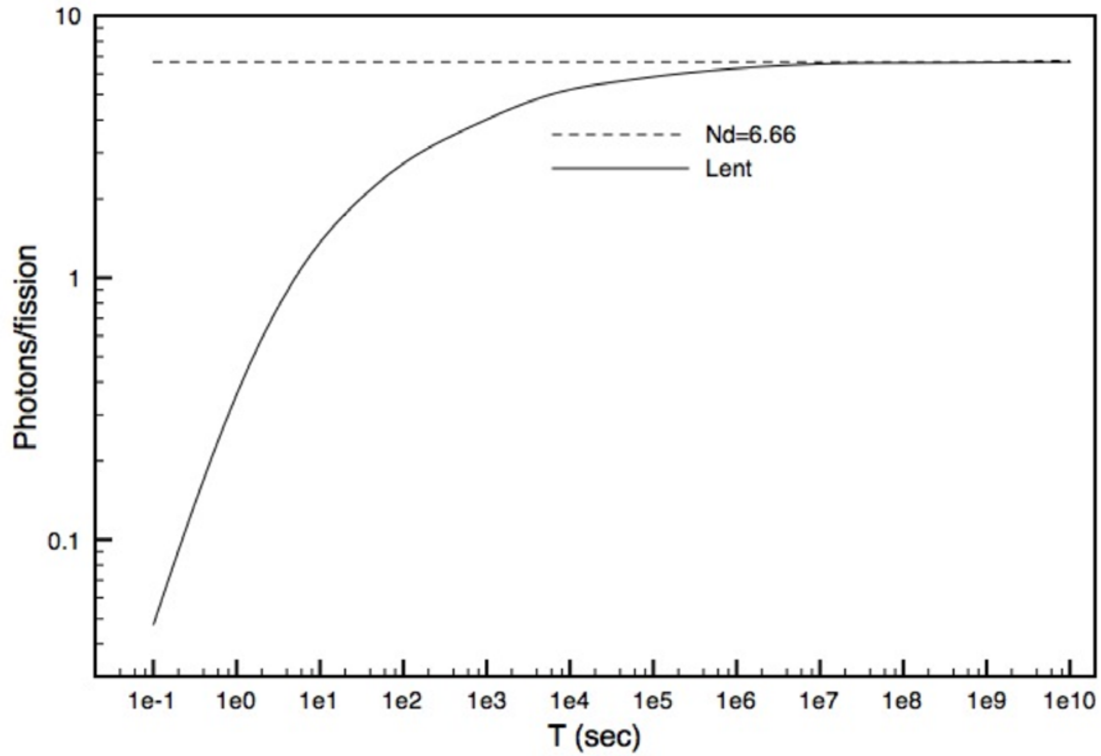
Results (1)

Lent's calculations produce delayed FP gamma multiplicities about 4% lower than Yanagisawa's results **– good agreement!**



Results (2)

Lent performed FP decay calculations out to 1e10 seconds and integrated the data to yield the photons/fission in the time interval [0, T]:



Results (3)

Ed Lent compared his calculated multiplicity against measured data …

Godiva

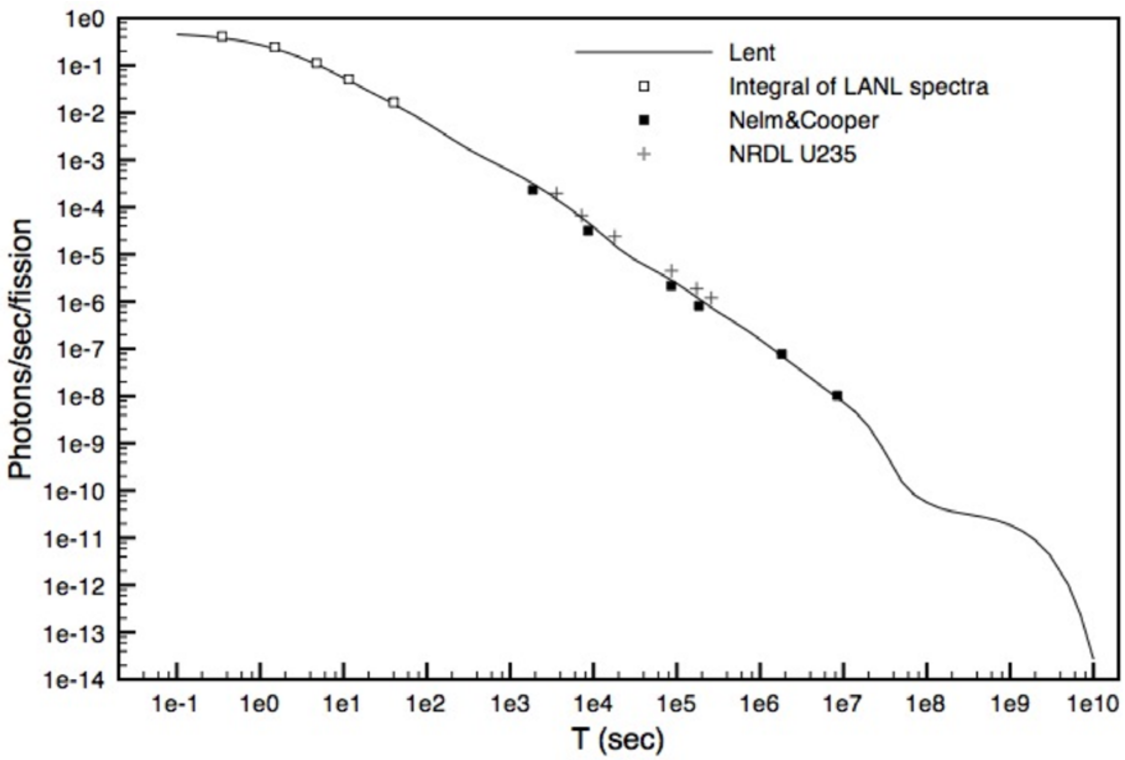
LAMS-2642

Nelms & Cooper

Health Physics, **1**, 427, 1959

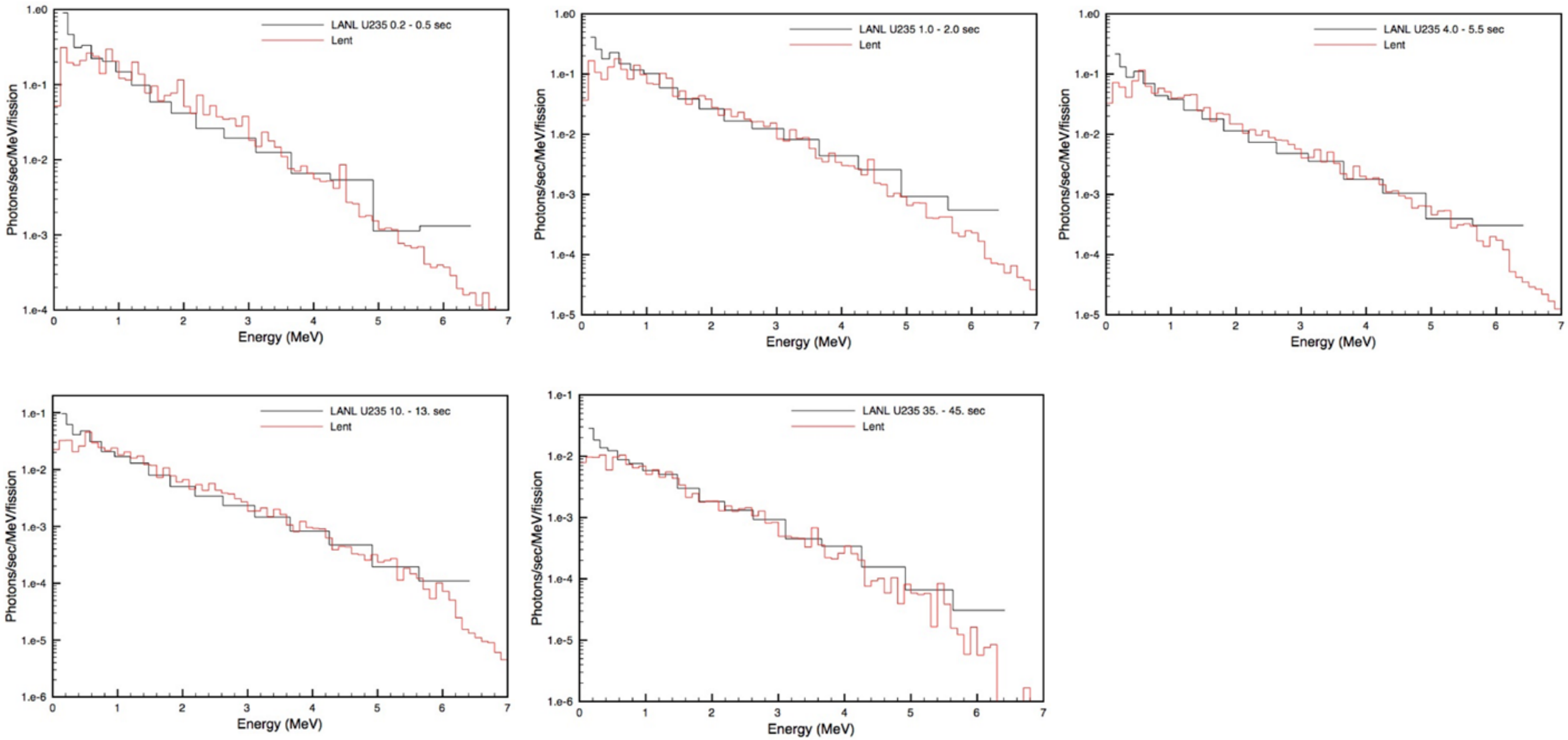
NRDL

NSE, **29**, 432, 1967



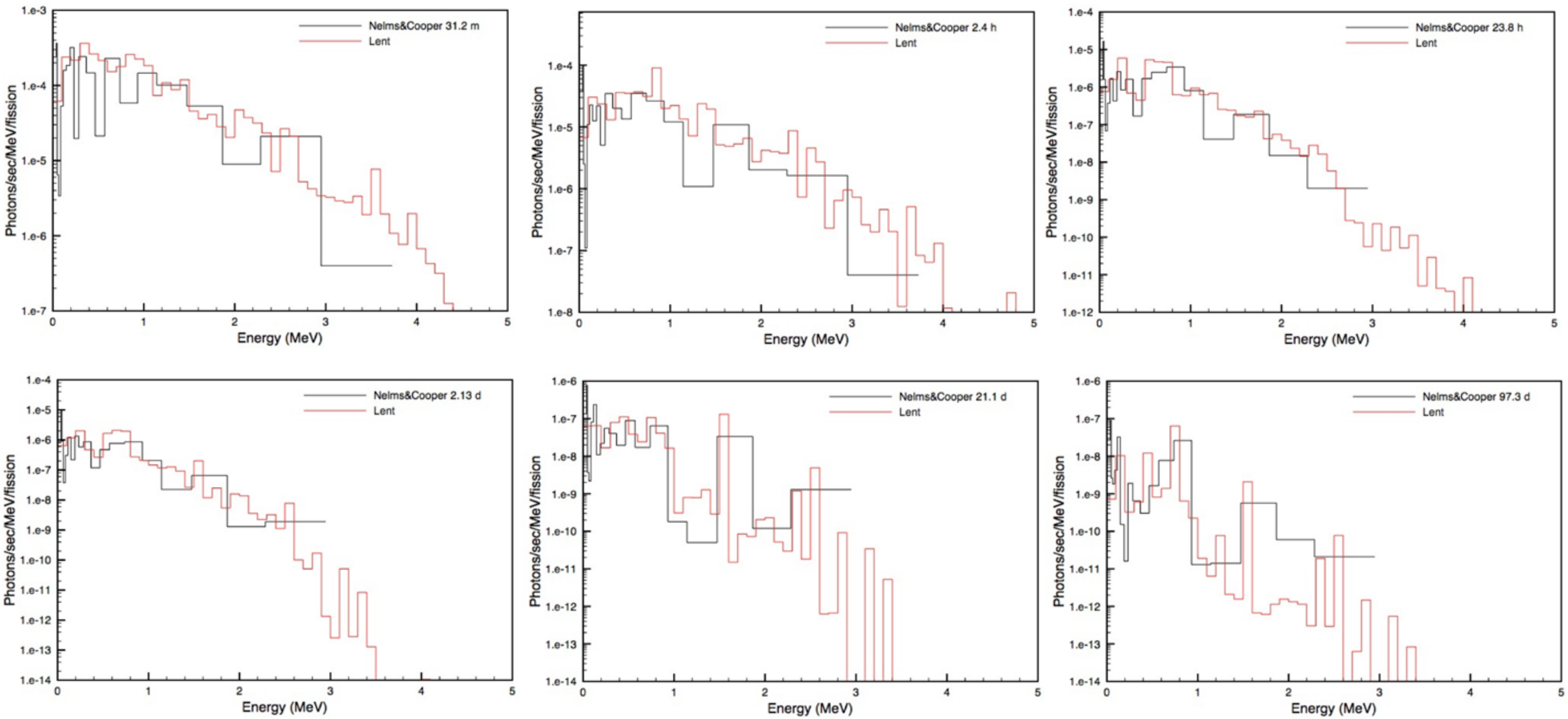
Results (4)

… and Ed Lent compared his calculated spectra against LANL (Godiva) measured data at early times (< 1 min) …



Results (5)

… and Ed Lent compared his calculated <u>spectra</u> against Nelms & Cooper measured data later times (1/2 hour to 97 days) …

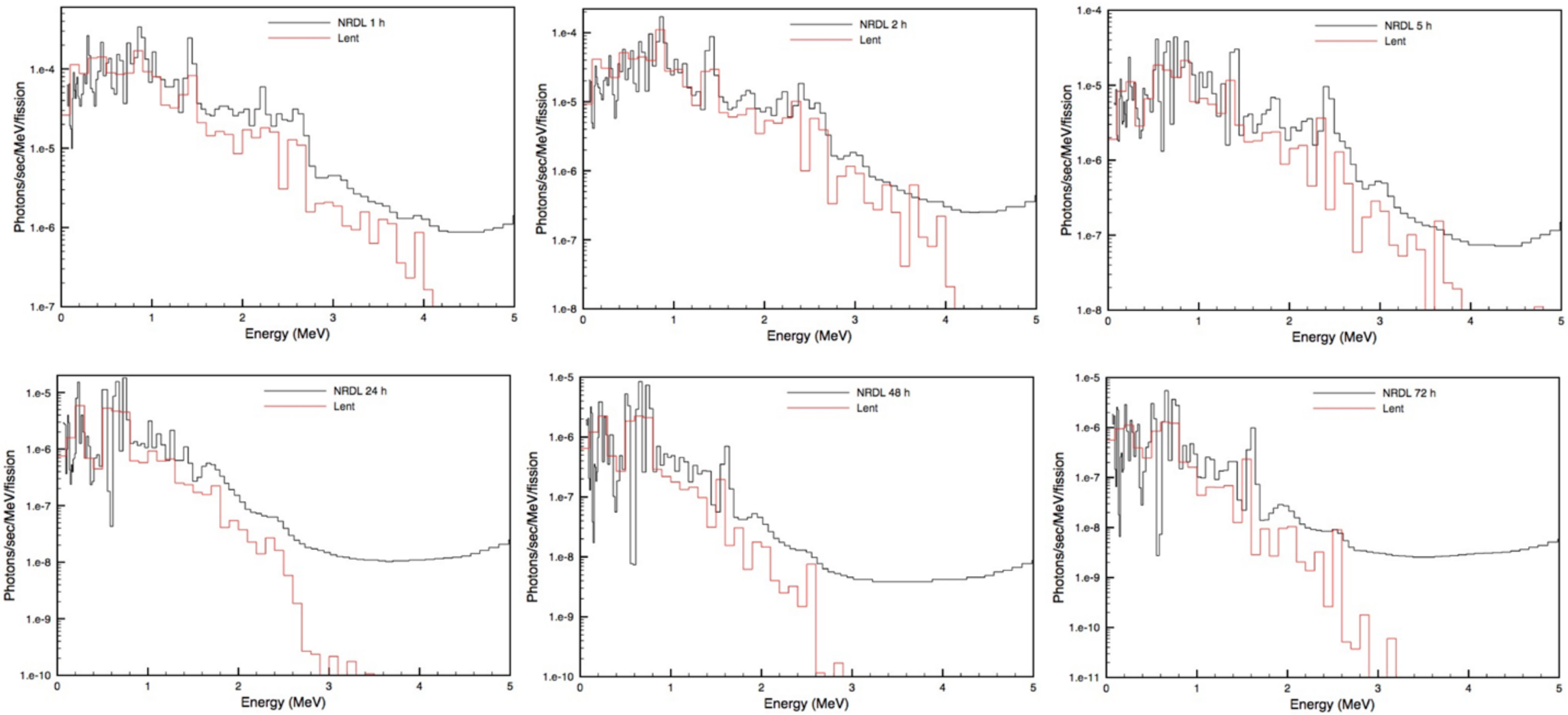


Results (6)

Note:
Lent > NRDL (E > 2 MeV)

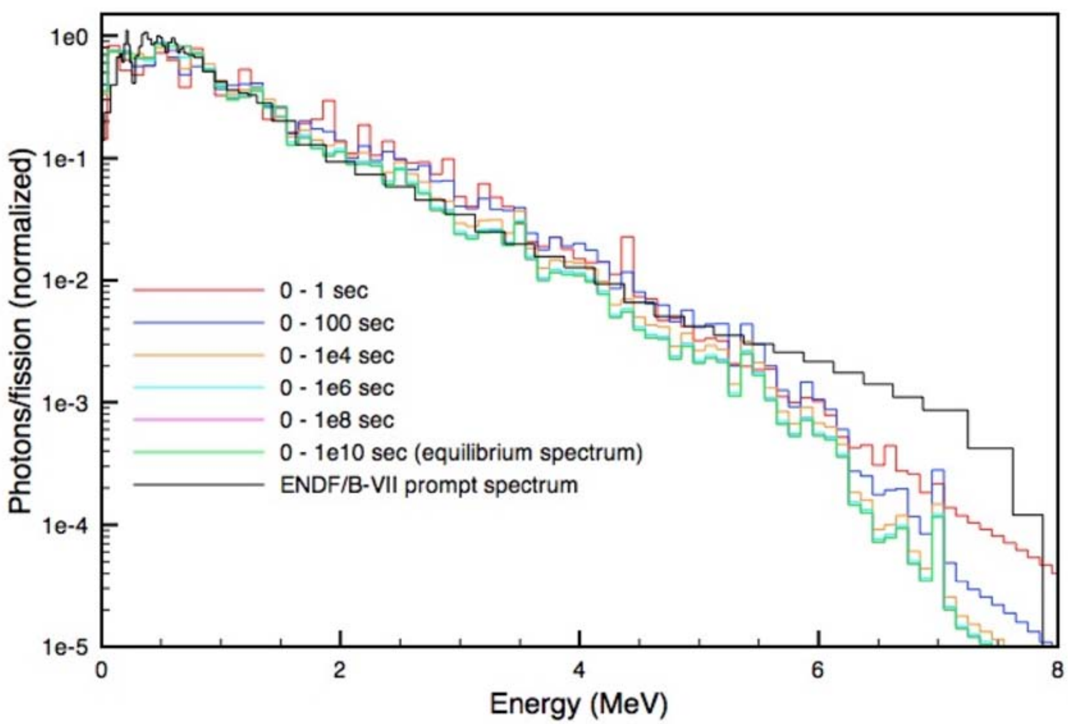
Recall:
Lent ≈ LANL (Slide 11)
Lent ≈ Nelms & Cooper (Slide 12)

… and Ed Lent compared his calculated <u>spectra</u> against NRDL measured data late times ….



Results (7) and Conclusions

Ed Lent also normalized and compared calculated spectra for various times ….
and compared it to the prompt spectrum …



1

(a) Use of an equilibrium delayed photon spectrum is superior to the current shielding practice of using the prompt spectrum to represent delayed photons.

(b) Time-dependent multiplicity data with equilibrium spectra is adequate for criticality accident "slide-rule" type calculations (see slide 16).

(c) This data is proposed for inclusion in ENDF/B-VII.1.

(d) Average parameters (see slide 4):
 6.66 photons/fission
 6.22 MeV/fission
 0.934 MeV/photon

2

Time-dependent point (not shown) and binned spectra are also available.

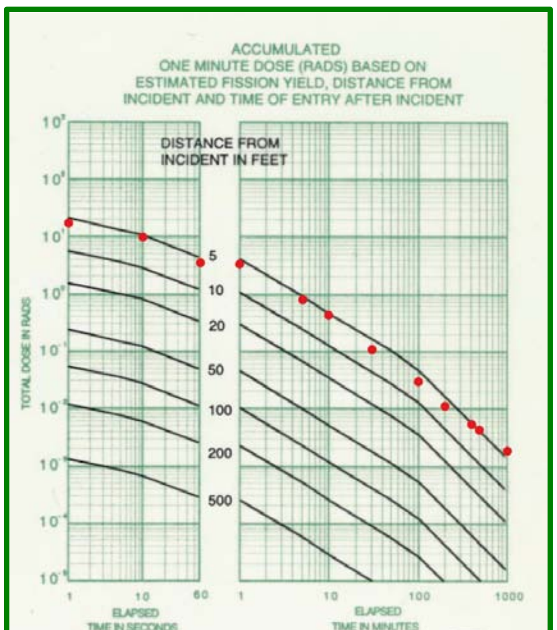
3

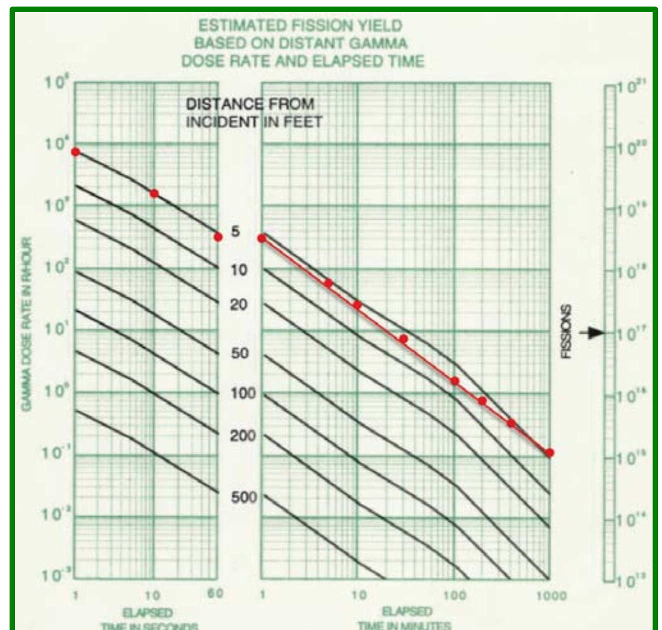
Time-dependent line and continuous spectra (not shown) are available.

Criticality accident slide-rule

Preliminary work by Heinrichs using COG11x demonstrates good agreement with the ORNL "slide rule" using: (a) Yanasigawa's time-dependent photon multiplicity data and (b) equilibrium prompt gamma spectrum.

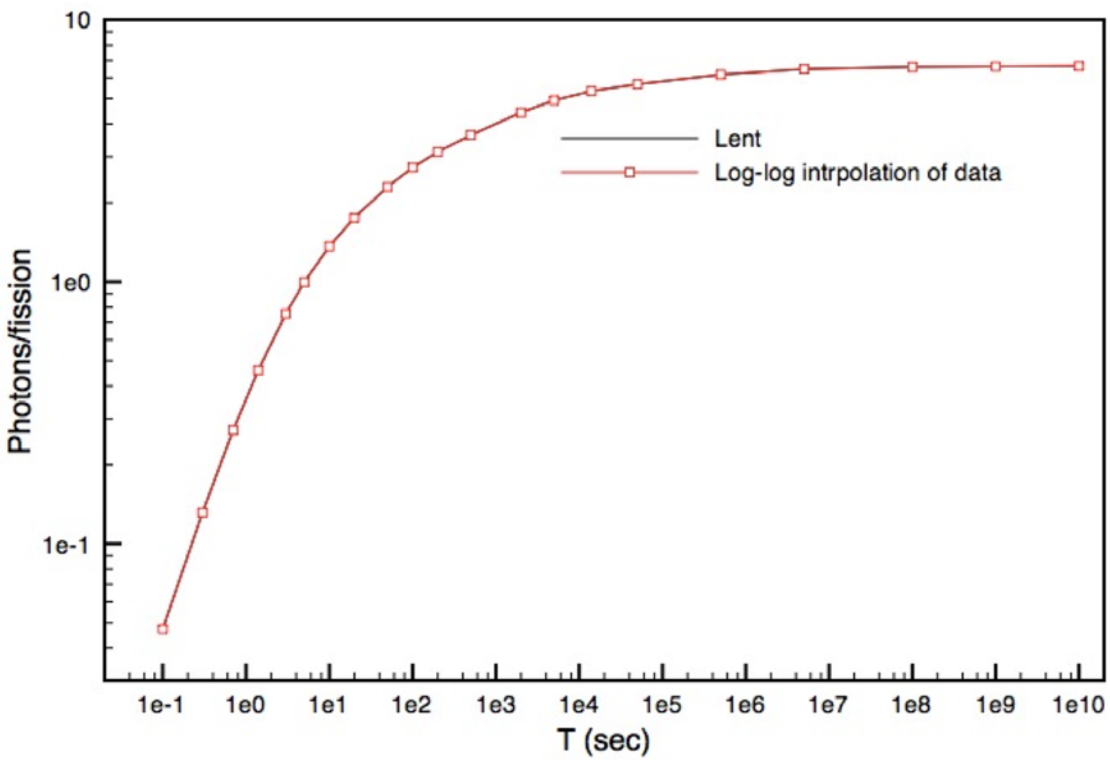
These calculations will be repeated with Lent's time-dependent multiplicity and equilibrium delayed spectrum for ^{235}U proposed for inclusion in ENDF/B-VII.1. Similar data for ^{238}U needed.





ENDF/B-VII.1 Proposed Multiplicity Data

T(sec)	Nd(photons/fission)
1.e-1	0.047
3.e-1	0.131
7.e-1	0.271
1.4e0	0.459
3.e0	0.754
5.e0	0.997
1.e1	1.364
2.e1	1.754
5.e1	2.301
1.e2	2.728
2.e2	3.131
5.e2	3.626
2.e3	4.426
5.e3	4.924
1.4e4	5.344
5.e4	5.675
5.e5	6.175
5.e6	6.487
1.e8	6.610
1.e9	6.634
1.e10	6.659



The plot (above) shows N_d vs T as a solid black line and the 21 points from the table (on the left) with a log-log interpolation between points as a red line with squares. A 21 point log-log table accurately represents the data.

Proposed ENDF/B Manual Changes

Proposed Changes to ENDF/B Manual

MF = 1 Changes:

Page 1.1, General Information:

"File 1 may contain up to four ..." changed to "File 1 may contain up to five additional sections giving fission neutron and photon yields and energy release information."

New Section:

Delayed Photon Data (MT=xxx)

This section describes the delayed photon/fission versus time.

Formats

Discrete Representation (LNU=2)

The following quantities are defined:

NR, NP, t_{int} Standard TAB1 parameters.

$M_d(t)$ Delayed photons/fission.

The structure of the time dependence data block is:

[MAT, 1, xxx/ ZA, AWR, 0, LNU=2, 0, 0] HEAD

[MAT, 1, xxx/ 0.0, 0.0, 0, 0, NR, NP/t_{int}/$M_d(t)$] TAB1

[MAT, 1, 0/ 0.0, 0.0, 0, 0, 0, 0] SEND

The photon energy spectra versus time may be entered in the MF=15 section as usual with MT=xxx.

ENDF/B-VII.1 – MF1 Data

Fast neutron induced fission – time-dependent delayed photon multiplicity (see slide 17)

```
92235.0000 233.024800          0          2          0          09228 1xxx  nnn
0.0        0.0                 0          0          2          229228 1xxx  nnn
           2          2          22          5                   9228 1xxx  nnn
0.00000000 0.00000000 0.10000000 0.04722000 0.30000000 0.131500009228 1xxx  nnn
0.70000000 0.27110000 1.40000000 0.45920000 3.00000000 0.753900009228 1xxx  nnn
5.00000000 0.99690000 10.0000000 1.36400000 20.0000000 1.754000009228 1xxx  nnn
50.0000000 2.30100000 100.000000 2.72800000 200.000000 3.131000009228 1xxx  nnn
500.000000 3.62600000 2000.00000 4.42600000 5000.00000 4.924000009228 1xxx  nnn
14000.0000 5.34400000 50000.0000 5.67500000 500000.000 6.175000009228 1xxx  nnn
5000000.00 6.48700000 100000000. 6.61000000 1.000000+9 6.634000009228 1xxx  nnn
1.00000+10 6.65900000                                             9228 1  0  nnn
```

ENDF/B-VII.1 – MF15 Data

Fast neutron induced fission – equilibrium delayed photon spectrum

```
92235.0000 233.024800          0          2          0         09228 1xxx  nnn
0.0        0.0                 0          0          2        229228 1xxx  nnn
        2          2          22          5                    9228 1xxx  nnn
0.00000000 0.00000000 0.10000000 0.04722000 0.30000000 0.131500009228 1xxx  nnn
0.70000000 0.27110000 1.40000000 0.45920000 3.00000000 0.753900009228 1xxx  nnn
5.00000000 0.99690000 10.0000000 1.36400000 20.0000000 1.754000009228 1xxx  nnn
50.0000000 2.30100000 100.000000 2.72800000 200.000000 3.131000009228 1xxx  nnn
500.000000 3.62600000 2000.00000 4.42600000 5000.00000 4.924000009228 1xxx  nnn
14000.0000 5.34400000 50000.0000 5.67500000 500000.000 6.175000009228 1xxx  nnn
5000000.00 6.48700000 100000000. 6.61000000 1.000000+9 6.634000009228 1xxx  nnn
1.00000+10 6.65900000                                  9228 1  0  nnn


[Replace data on slide 20 with…]

92235.0000 233.024800          0          0          1         0922815xxx  nnn
0.000000+0 0.000000+0          0          1          1         2922815xxx  nnn
        2          2                                  922815xxx  nnn
1.000000-5 1.000000+0 2.000000+7 1.000000+0                     922815xxx  nnn
0.000000+0 0.000000+0          0          0          1         2922815xxx  nnn
        2          2                                  922815xxx  nnn
0.000000+0 1.000000-5          0          0          1        91922815xxx  nnn
       91          1                                  922815xxx  nnn
0.000000+0 3.458830-7 1.000000+5 7.434270-7 2.000000+5 7.058280-7922815xxx  nnn
3.000000+5 6.386830-7 4.000000+5 6.408820-7 5.000000+5 8.406810-7922815xxx  nnn
6.000000+5 8.044810-7 7.000000+5 8.030020-7 8.000000+5 6.891050-7922815xxx  nnn
9.000000+5 5.072430-7 1.000000+6 3.795330-7 1.100000+6 2.936460-7922815xxx  nnn
1.200000+6 3.101870-7 1.300000+6 3.485810-7 1.400000+6 2.525850-7922815xxx  nnn
1.500000+6 2.580830-7 1.600000+6 1.253590-7 1.700000+6 1.442600-7922815xxx  nnn
1.800000+6 1.180430-7 1.900000+6 1.025220-7 2.000000+6 1.104450-7922815xxx  nnn
2.100000+6 8.728850-8 2.200000+6 8.827700-8 2.300000+6 8.471720-8922815xxx  nnn
2.400000+6 6.040160-8 2.500000+6 7.917830-8 2.600000+6 5.958810-8922815xxx  nnn
2.700000+6 4.974660-8 2.800000+6 3.639520-8 2.900000+6 3.499540-8922815xxx  nnn
3.000000+6 2.318510-8 3.100000+6 2.143040-8 3.200000+6 2.398240-8922815xxx  nnn
3.300000+6 2.438270-8 3.400000+6 1.893370-8 3.500000+6 2.825040-8922815xxx  nnn
3.600000+6 1.454330-8 3.700000+6 9.774890-9 3.800000+6 1.129920-8922815xxx  nnn
3.900000+6 1.092020-8 4.000000+6 1.069410-8 4.100000+6 9.522600-9922815xxx  nnn
4.200000+6 7.546720-9 4.300000+6 4.865320-9 4.400000+6 5.398290-9922815xxx  nnn
4.500000+6 3.853190-9 4.600000+6 3.321950-9 4.700000+6 3.253600-9922815xxx  nnn
4.800000+6 2.235920-9 4.900000+6 2.847040-9 5.000000+6 2.065120-9922815xxx  nnn
5.100000+6 2.272280-9 5.200000+6 2.112090-9 5.300000+6 1.097190-9922815xxx  nnn
5.400000+6 2.460170-9 5.500000+6 1.645790-9 5.600000+6 1.018000-9922815xxx  nnn
5.700000+6 6.48741-10 5.800000+6 5.17808-10 5.900000+6 7.03085-10922815xxx  nnn
6.000000+6 5.22423-10 6.100000+6 4.86379-10 6.200000+6 3.50142-10922815xxx  nnn
6.300000+6 1.42181-10 6.400000+6 1.22288-10 6.500000+6 7.06204-11922815xxx  nnn
6.600000+6 7.72199-11 6.700000+6 9.17392-11 6.800000+6 4.68845-11922815xxx  nnn
6.900000+6 3.38663-11 7.000000+6 1.13724-10 7.100000+6 1.96341-11922815xxx  nnn
7.200000+6 1.37558-11 7.300000+6 1.17941-11 7.400000+6 1.02336-11922815xxx  nnn
7.500000+6 8.82683-12 7.600000+6 7.54421-12 7.700000+6 6.38198-12922815xxx  nnn
7.800000+6 5.32734-12 7.900000+6 4.36959-12 8.000000+6 3.49494-12922815xxx  nnn
8.100000+6 2.73127-12 8.200000+6 2.42008-12 8.300000+6 2.13654-12922815xxx  nnn
8.400000+6 1.87794-12 8.500000+6 1.64239-12 8.600000+6 1.42930-12922815xxx  nnn
8.700000+6 1.23689-12 8.800000+6 1.06512-12 8.900000+6 9.33572-13922815xxx  nnn
9.000000+6 0.000000+0                                  922815xxx  nnn
0.000000+0 2.000000+7          0          0          1        91922815xxx  nnn
       91          1                                  922815xxx  nnn
0.000000+0 3.458830-7 1.000000+5 7.434270-7 2.000000+5 7.058280-7922815xxx  nnn
3.000000+5 6.386830-7 4.000000+5 6.408820-7 5.000000+5 8.406810-7922815xxx  nnn
6.000000+5 8.044810-7 7.000000+5 8.030020-7 8.000000+5 6.891050-7922815xxx  nnn
9.000000+5 5.072430-7 1.000000+6 3.795330-7 1.100000+6 2.936460-7922815xxx  nnn
1.200000+6 3.101870-7 1.300000+6 3.485810-7 1.400000+6 2.525850-7922815xxx  nnn
1.500000+6 2.580830-7 1.600000+6 1.253590-7 1.700000+6 1.442600-7922815xxx  nnn
1.800000+6 1.180430-7 1.900000+6 1.025220-7 2.000000+6 1.104450-7922815xxx  nnn
2.100000+6 8.728850-8 2.200000+6 8.827700-8 2.300000+6 8.471720-8922815xxx  nnn
2.400000+6 6.040160-8 2.500000+6 7.917830-8 2.600000+6 5.958810-8922815xxx  nnn
2.700000+6 4.974660-8 2.800000+6 3.639520-8 2.900000+6 3.499540-8922815xxx  nnn
3.000000+6 2.318510-8 3.100000+6 2.143040-8 3.200000+6 2.398240-8922815xxx  nnn
3.300000+6 2.438270-8 3.400000+6 1.893370-8 3.500000+6 2.825040-8922815xxx  nnn
3.600000+6 1.454330-8 3.700000+6 9.774890-9 3.800000+6 1.129920-8922815xxx  nnn
3.900000+6 1.092020-8 4.000000+6 1.069410-8 4.100000+6 9.522600-9922815xxx  nnn
4.200000+6 7.546720-9 4.300000+6 4.865320-9 4.400000+6 5.398290-9922815xxx  nnn
4.500000+6 3.853190-9 4.600000+6 3.321950-9 4.700000+6 3.253600-9922815xxx  nnn
4.800000+6 2.235920-9 4.900000+6 2.847040-9 5.000000+6 2.065120-9922815xxx  nnn
5.100000+6 2.272280-9 5.200000+6 2.112090-9 5.300000+6 1.097190-9922815xxx  nnn
5.400000+6 2.460170-9 5.500000+6 1.645790-9 5.600000+6 1.018000-9922815xxx  nnn
5.700000+6 6.48741-10 5.800000+6 5.17808-10 5.900000+6 7.03085-10922815xxx  nnn
6.000000+6 5.22423-10 6.100000+6 4.86379-10 6.200000+6 3.50142-10922815xxx  nnn
6.300000+6 1.42181-10 6.400000+6 1.22288-10 6.500000+6 7.06204-11922815xxx  nnn
6.600000+6 7.72199-11 6.700000+6 9.17392-11 6.800000+6 4.68845-11922815xxx  nnn
6.900000+6 3.38663-11 7.000000+6 1.13724-10 7.100000+6 1.96341-11922815xxx  nnn
7.200000+6 1.37558-11 7.300000+6 1.17941-11 7.400000+6 1.02336-11922815xxx  nnn
7.500000+6 8.82683-12 7.600000+6 7.54421-12 7.700000+6 6.38198-12922815xxx  nnn
7.800000+6 5.32734-12 7.900000+6 4.36959-12 8.000000+6 3.49494-12922815xxx  nnn
8.100000+6 2.73127-12 8.200000+6 2.42008-12 8.300000+6 2.13654-12922815xxx  nnn
8.400000+6 1.87794-12 8.500000+6 1.64239-12 8.600000+6 1.42930-12922815xxx  nnn
8.700000+6 1.23689-12 8.800000+6 1.06512-12 8.900000+6 9.33572-13922815xxx  nnn
9.000000+6 0.000000+0                                  922815xxx  nnn
                                    922815  0  nnn
```

Summary

* Delayed photon multiplicity and spectra are available for ^{235}U for thermal, fast, fusion neutron induced fission.

* Time-dependent multiplicity and time-independent (equilibrium) spectra are proposed for inclusion in ENDF/B-VII.1 – useful in assessing criticality accident doses.

* Are time-dependent point or binned spectra needed for more accurate calculations or other applications?

* Is there interest in time-dependent line and continuous spectra for use in identifying specific fission product signatures?

* Work on ^{239}Pu in progress.

Questions?

* This method can be applied to spontaneous fission of U238, Cm244, Cm246, Cm248, Cf250, Cf252, Es253, Fm254, and Fm256; thermal neutron induced fission of Th227, Th229, U232, U233, U235, Np237, Pu239, Pu240, Pu241, Pu242, Am241, Am242, Cm245, Cf249, Cf251, Es254, and Fm255; the fast neutron induced fission of Pa231, Th232, U233, U234, U235, U236, U237, U238, Np237, Np238, Pu238, Pu239, Pu240, Pi241, Pu242, Am241, Am243, Cm242, Cm243, Cm244, Cm246, and Cm248; and high energy neutron induced fission of Th232, U233, U234, U235, U236, U238, Np237, Pu239, Pu240, Pu242, and Am241.