

# Lawrence Livermore National Laboratory

## Delayed Fission Gammas



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# Introduction

**LLNL had a DOE NCSP task to test the suitability of ENDF/B-VII.0 delayed fission gamma 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

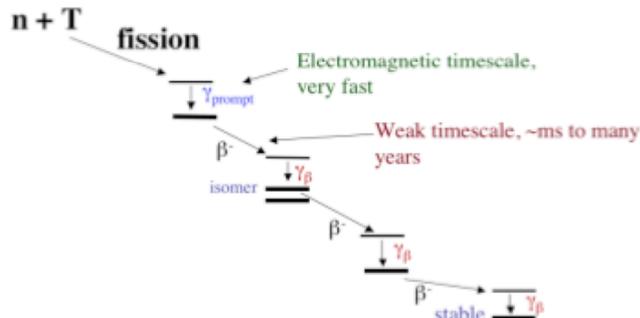
# ENDF/B-VII.0

ENDF/B-VII.0 data provided by LLNL

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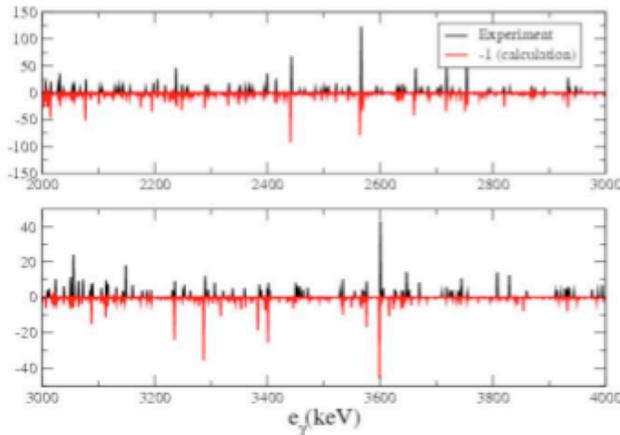


## Simulating $\beta$ -delayed $\gamma$ 's from fission



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

- Generate fission fragment from England and Rider. ← Monte Carlo
- Follow  $\beta$ -decay chain to stability ← Monte Carlo
- Collect  $\gamma$ 's along the way. ← NuDat database
- Generally good agreement w/ experiment of Norman et al.



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

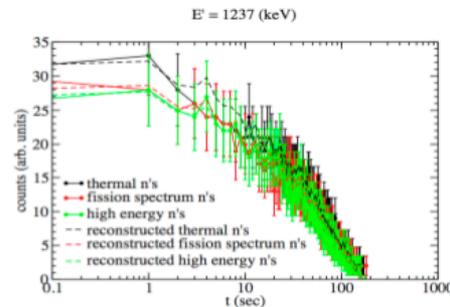
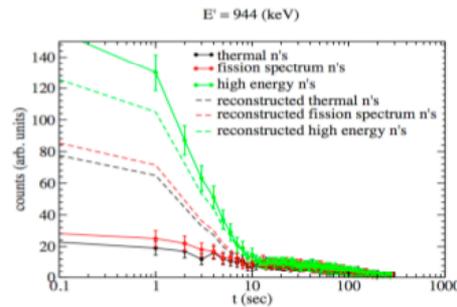
UCRL-PRES-225819

## Putting data in ENDF/B format

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- Approximate  $\gamma$  spectrum/unit time as product of time distribution and multiplicity:
- $s_{\gamma}(E, E_{\gamma}, t) = y(E, E_{\gamma})T(t)$
- In MT=460, MF=1,14
- 3129 lines in  $^{239}\text{Pu}$ , 3262 lines in  $^{235}\text{U}$
- Data in use in COG transport code



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Unfortunately, Pruet data way off in energy and multiplicity; e.g., for  $^{235}\text{U}$ :

<u>Nd</u>	<u>Ed</u>	<u>Reference</u>
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		PhysRevC 6:1023 (1967)

$Ed/Nd = 0.80$  MeV/g (Pruet) when it should be  $\sim 1$  MeV/g.

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

**This was a surprise!**

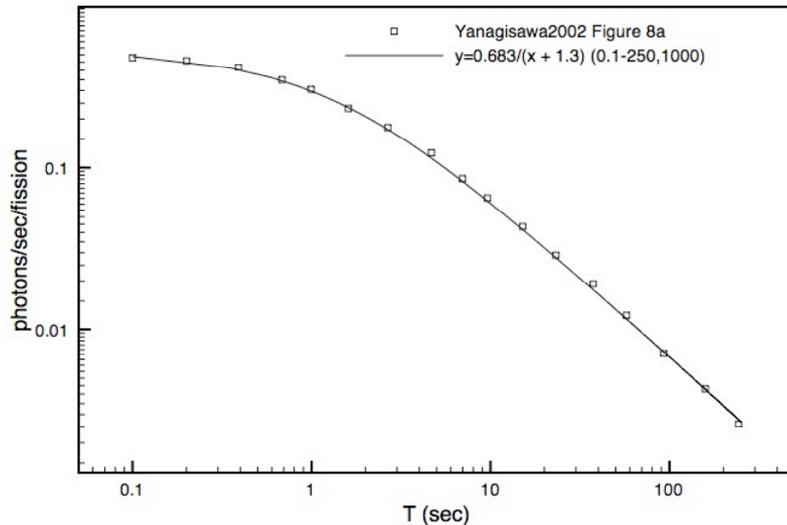
# Yanagisawa 2002

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

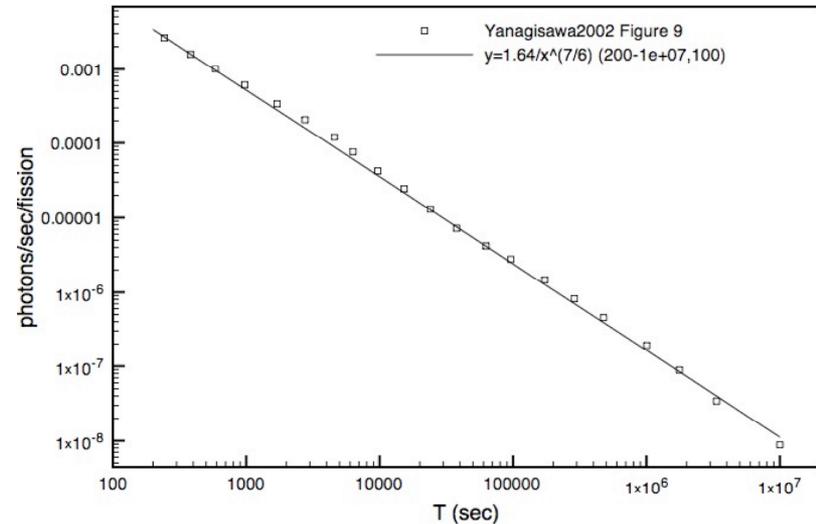
Way-Wigner  $T^{-1.2}$

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

$$n_d = 0.683/(T + 1.3), \quad T < 200 \text{ sec}$$



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



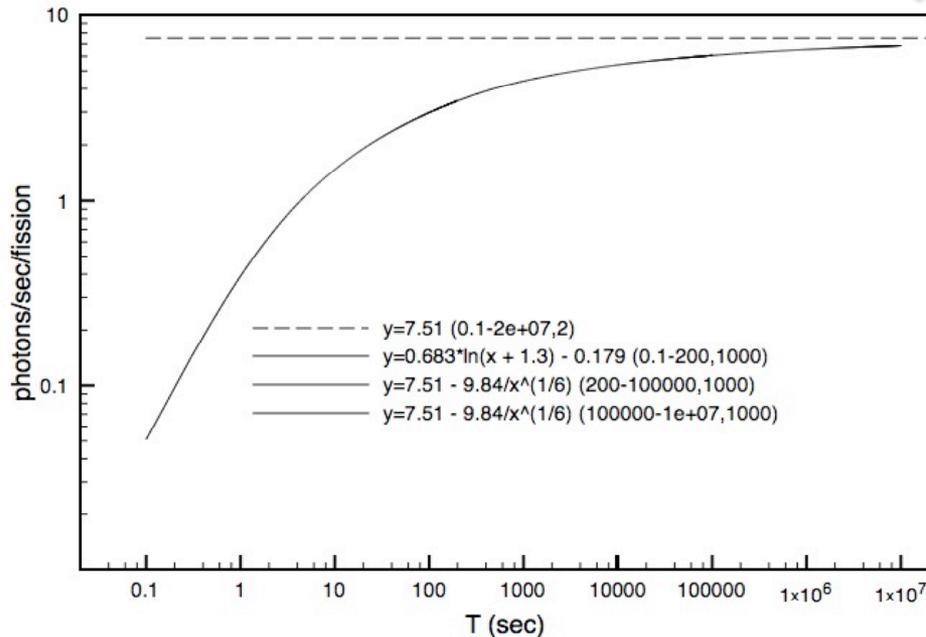
# Yanagisawa 2002

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  $[0, T]$ :

$$N_d = 0.683 \ln(T + 1.3) - 0.179, \quad T < 200 \text{ sec}$$

$$N_d = 7.51 - 9.84/T^{1/6}, \quad T > 200 \text{ sec}$$



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

Other researchers report lower values.

# LLNL DFG Accomplishments

Ed Lent replicated previous work by Pruett and Yanagisawa by developing new libraries and codes:

## COGFY

T.R. England and B.F. Rider database (LA-UR-94-3106, ENDF-349) of fission products (789 FP nuclides for  $^{235}\text{U}$  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

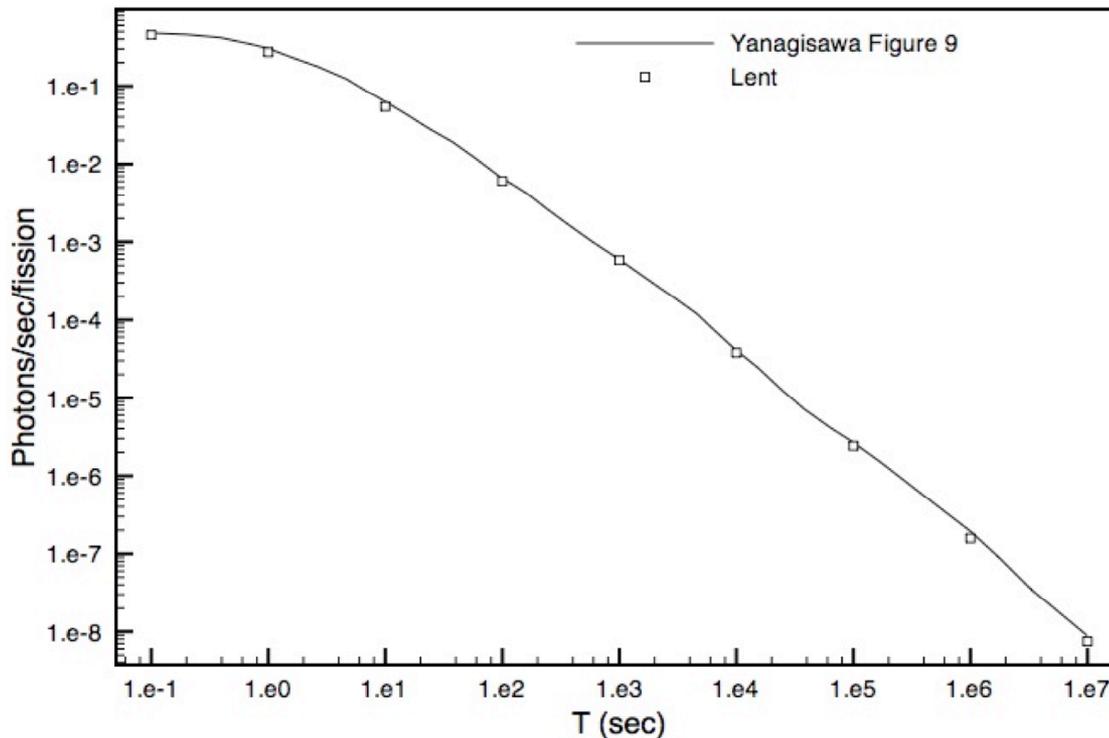
\* Bateman solutions previously developed for gamma emission from  $\alpha$ -decay

## 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 for  $^{235}\text{U}$  produce delayed FP gamma multiplicities about 4% lower than Yanagisawa's results – **good agreement!**

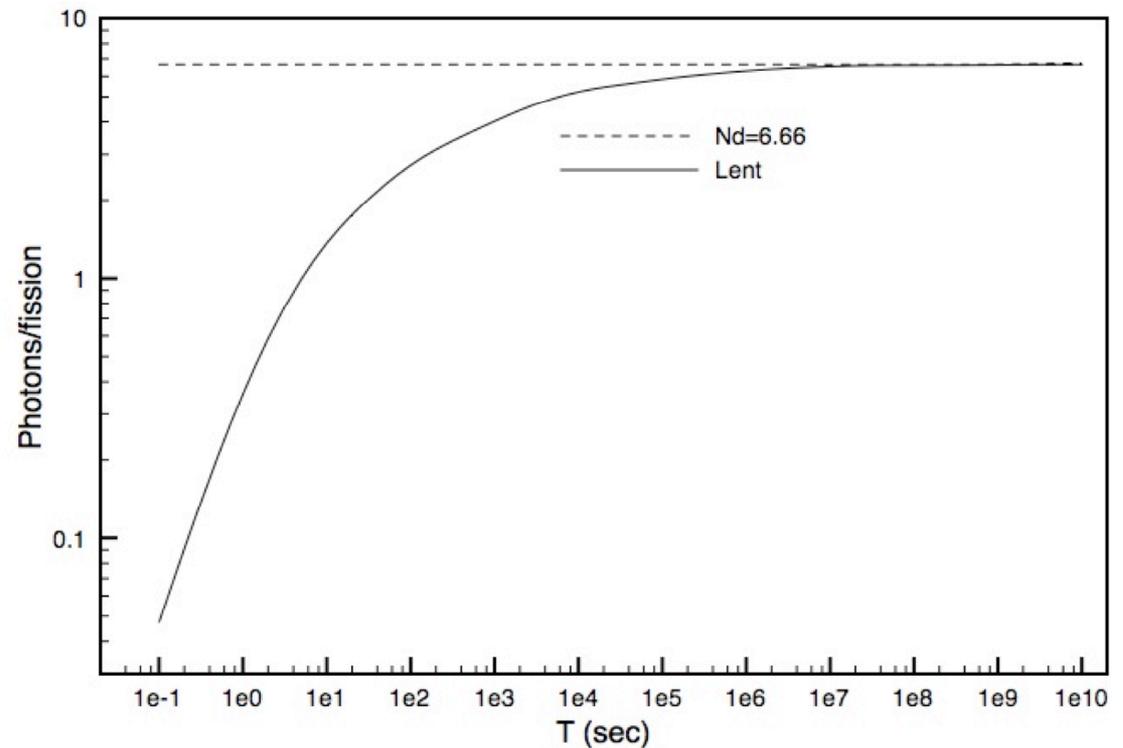


Using JENDL database instead of England & Rider may result in even better agreement.

JEFF data is also available.

# Results (2)

Lent performed FP decay calculations for  $^{235}\text{U}$  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 for  $^{235}\text{U}$  against measured data ...

## Godiva

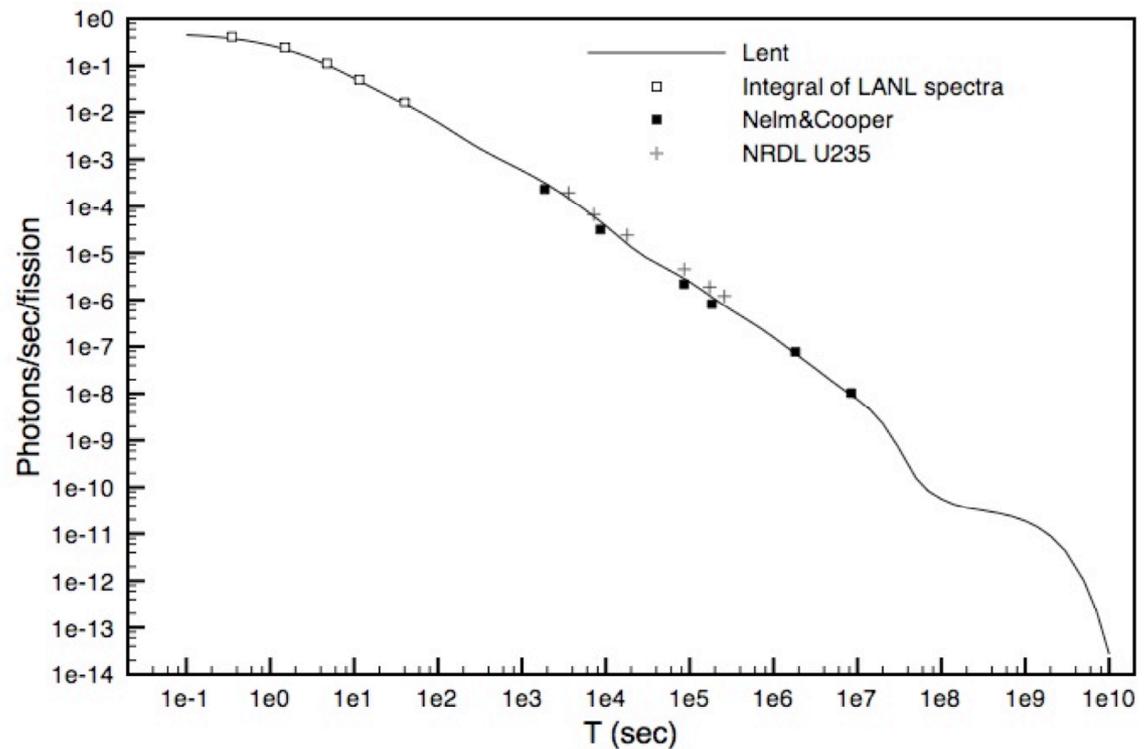
LAMS-2642

## Nelms & Cooper

Health Physics, 1, 427, 1959

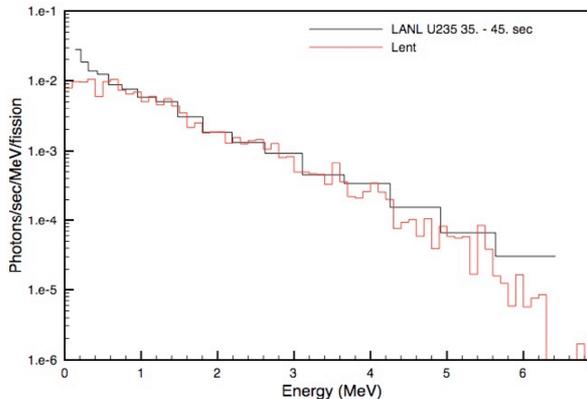
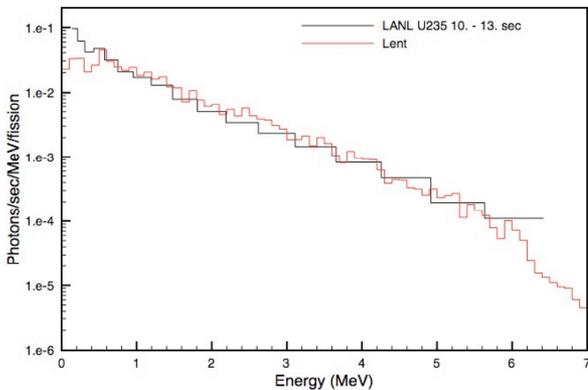
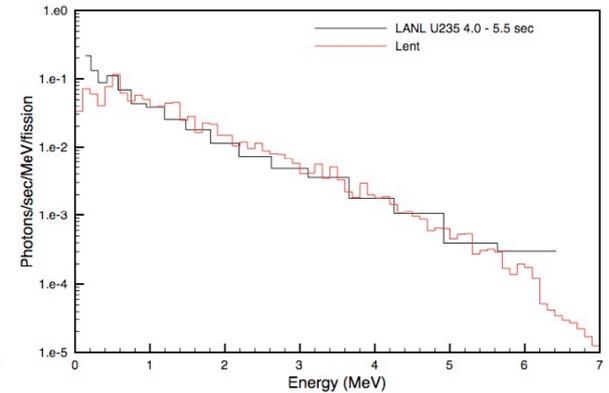
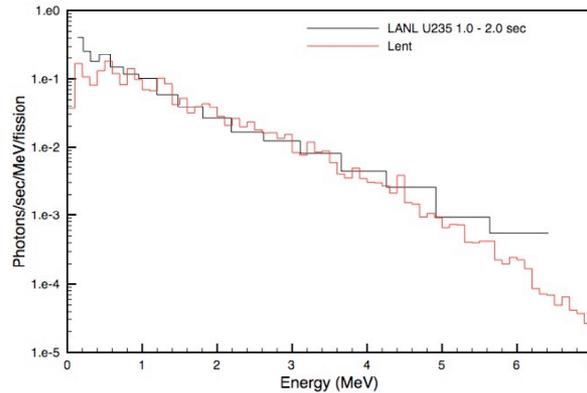
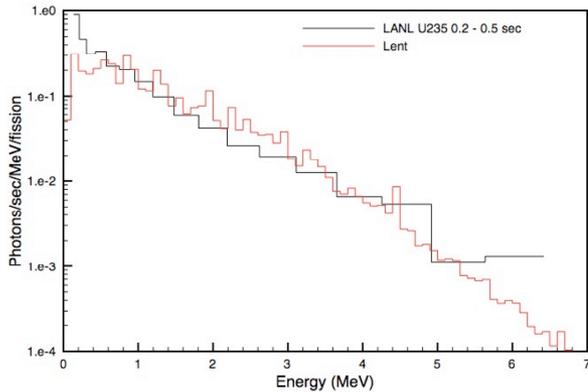
## NRDL

NSE, 29, 432, 1967



# Results (4)

... and Ed Lent compared his calculated spectra for  $^{235}\text{U}$  against LANL (Godiva) measured data at early times (< 1 min) ...

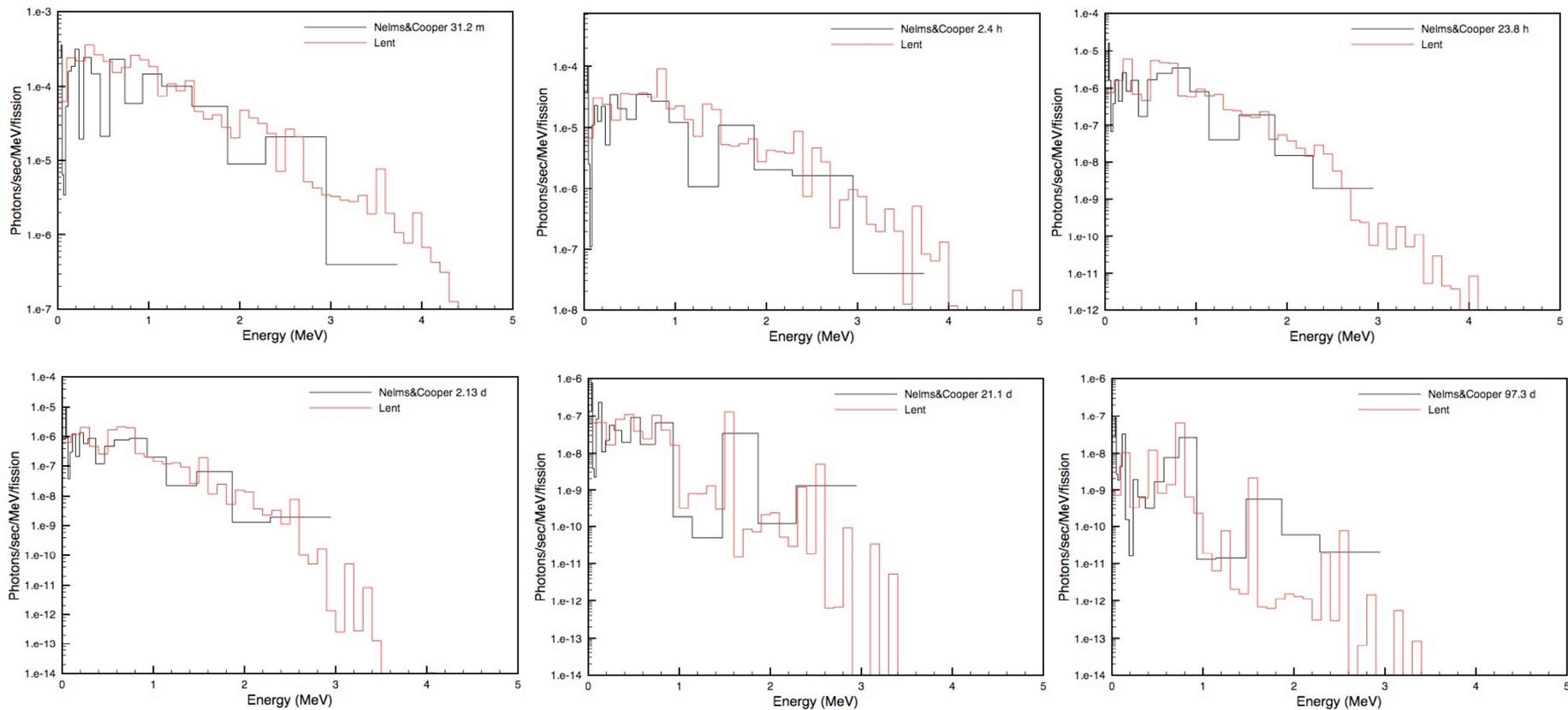


LANL and LLNL have submitted IERs for new Godiva foil activation measurements.

Modern gamma spectroscopy instruments should produce much better experimental spectra of benefit to NCSP in addition to NCT and RadChem.

# Results (5)

... and Ed Lent compared his calculated spectra for  $^{235}\text{U}$  against Nelms & Cooper measured data later times (1/2 hour to 97 days) ...



# Results (6)

... and Ed Lent compared his calculated spectra for  $^{235}\text{U}$  against NRDL measured data late times ....

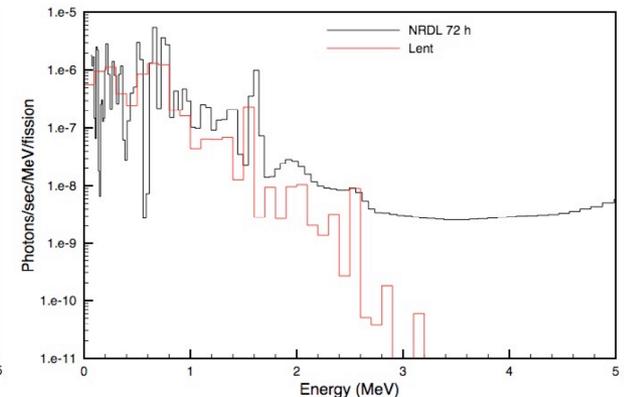
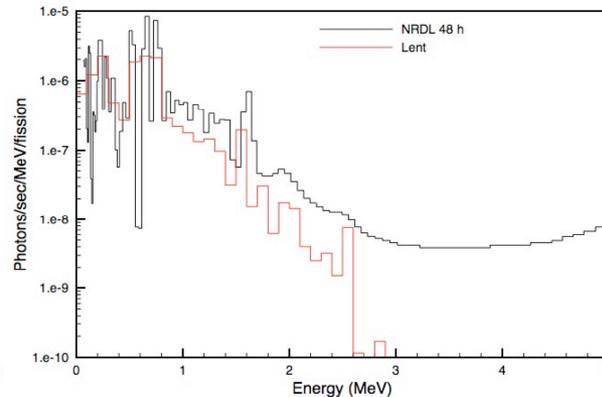
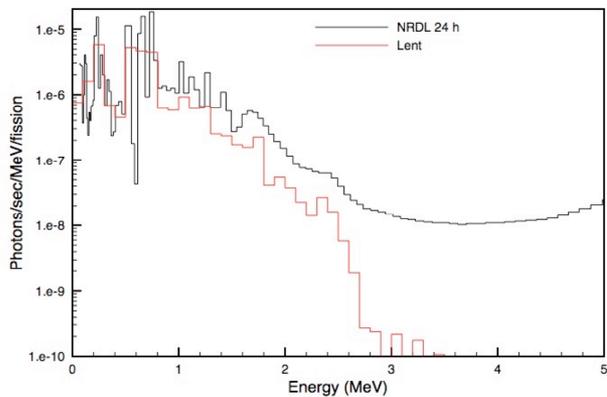
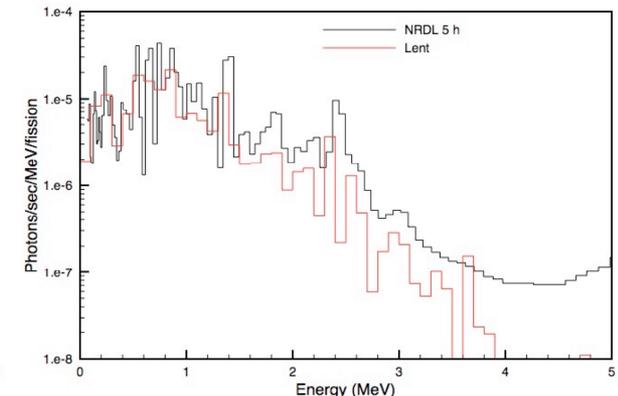
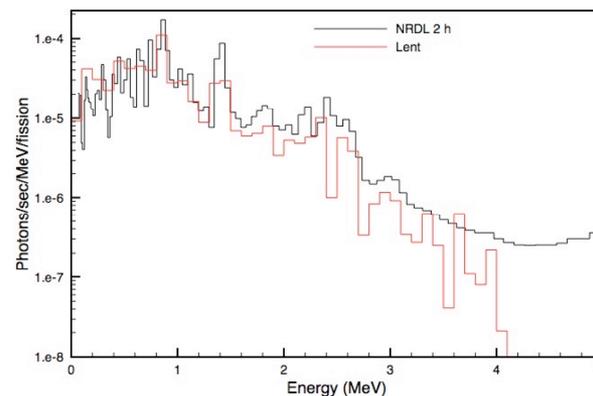
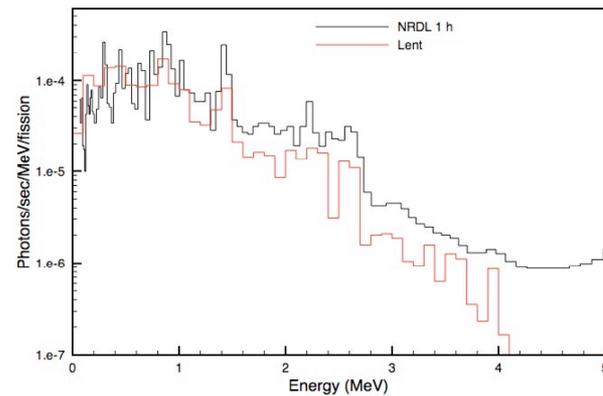
Note:

Lent > NRDL ( $E > 2$  MeV)

Recall:

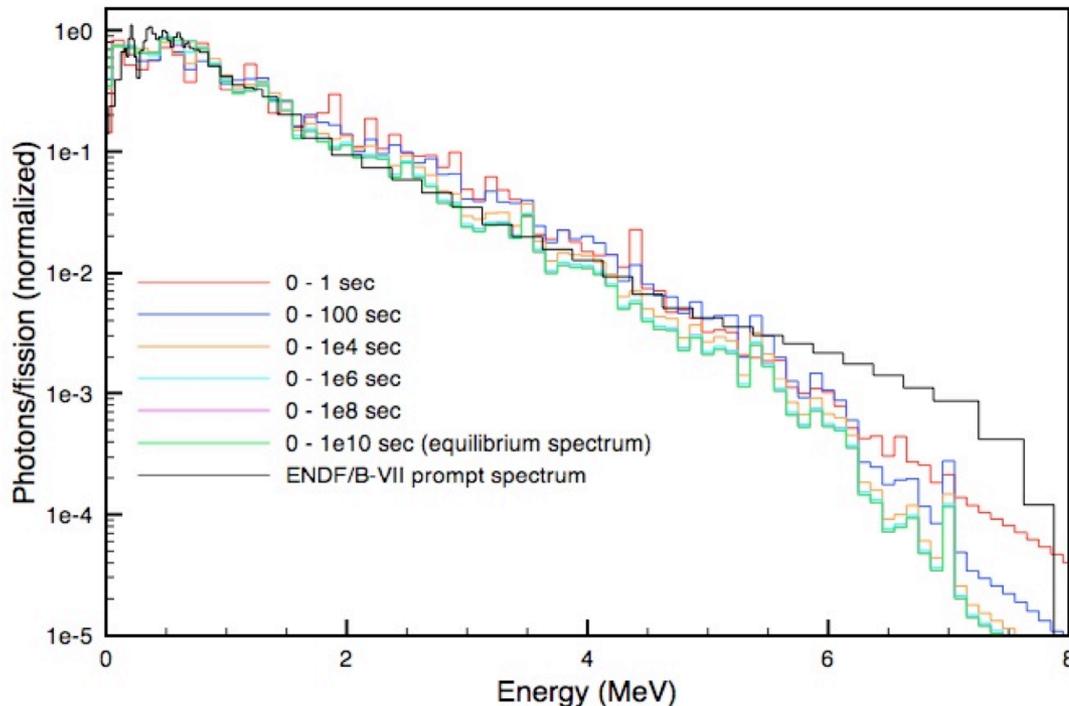
Lent  $\approx$  LANL (Slide 11)

Lent  $\approx$  Nelms & Cooper (Slide 12)



# Results (7) and Conclusions

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



1

- (a) Time-dependent multiplicity data with average spectra appear adequate for criticality accident “slide-rule” type calculations (see slide 16).
- (b) This data was proposed for inclusion in ENDF/B-VII at CSEWG (Nov 2010).
- (c) 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 (shown) are also available.

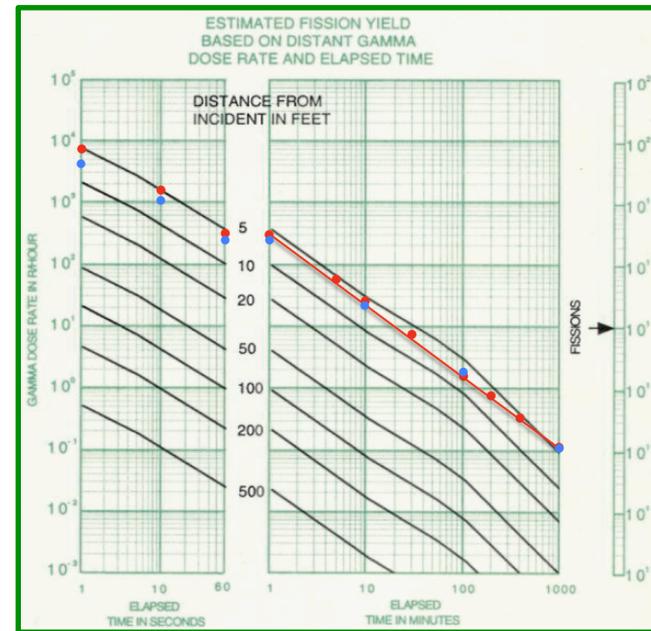
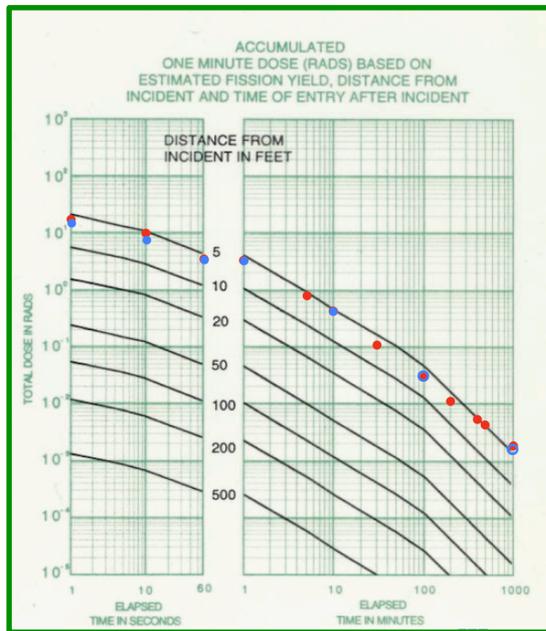
3

Time-dependent line and continuous spectra (not shown) are available. This data may be useful as a fission signature.

# Criticality accident slide-rule

Lent's results demonstrate good agreement with the ORNL "slide rule" using:

- COG11 using Yanasigawa's time-dependent  $^{235}\text{U}$  multiplicity data with prompt gamma spectrum
- COG11 using Lent's time-dependent  $^{234,235,238}\text{U}$  multiplicity and delayed gamma spectra
- Hand calculated dose (estimated from COG11DFG calculated rate at late times) (fix in progress)



# What's next?

- COGDFG library **completed** for fast neutron induced delayed fission gammas for:  
Th232, U233, U234, U235, U236, U238, Pu239, Pu240, Pu241.
- More **testing** needed (similar to U235 testing completed in 2010)
- Need COGDFG libraries for **thermal** neutron induced fission of:  
Th227, Th229, U232, U233, U235, Np237, Pu239, Pu240, Pu241, Pu242, Am241, Am242, Cm245, Cf249, Cf251, Es254, and Fm255;
- Need COGDFG libraries for the **fast** neutron induced fission of:  
Pa231, U237, Np237, Np238, Pu238, Pu242, Am241, Am243, Cm242, Cm243, Cm244, Cm246, and Cm248;
- Need COGDFG libraries for high energy (**14 MeV**) neutron induced fission of:  
Th232, U233, U234, U235, U236, U238, Np237, Pu239, Pu240, Pu242, and Am241;
- Need COGDFG libraries for **spontaneous fission** of:  
U238, Cm244, Cm246, Cm248, Cf250, Cf252, Es253, Fm254, and Fm256.
- **Need Criticality Slide Rule for Plutonium (Proposal)**